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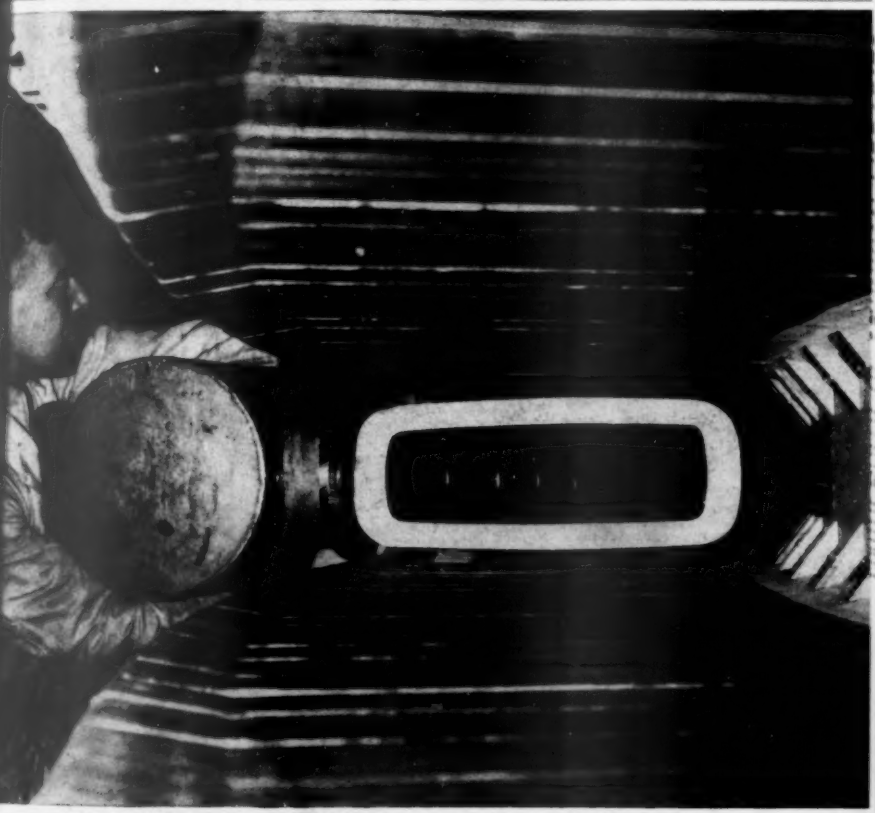

Vacuum box of Birmingham proton synchrotron; see pgs. 30, 97

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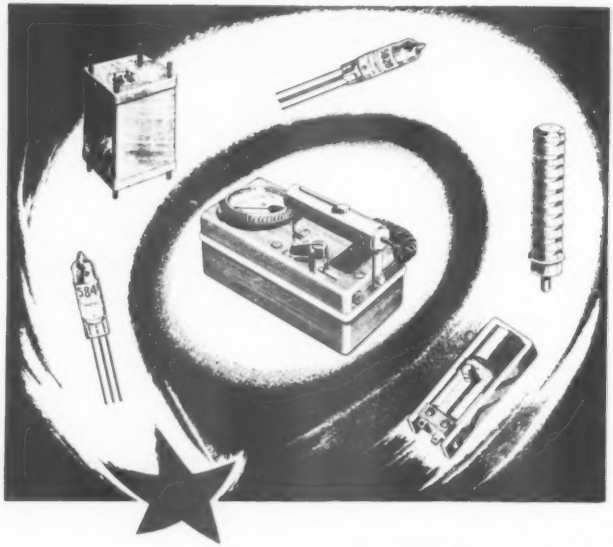

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GENERAL ELECTRIC

A Survey of Ceramics for Nuclear Reactors*

Tabulated and discussed here are the physical properties of twenty-four ceramic materials, with special emphasis on properties of interest in nuclear engineering. Possible ceramic applications in nuclear reactors and methods for preliminary evaluation of ceramic materials are outlined

By R. F. GELLER

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U. S. Department of Commerce, Washington, D. C.

THE CERAMIC ENGINEER who is interested in the development of materials for nuclear reactors but who, like the author, is not a nuclear physicist, and the nuclear physicist or engineer who may want to become more familiar with the physical and chemical properties of ceramics are the persons for whom this report was primarily written.

What Is Expected of Ceramics in Nuclear Reactors?

There are two classes of nuclear reactors or piles: homogeneous and heterogeneous. In the homogeneous reactor, the fuel, or fissionable material, is distributed uniformly throughout the active portion of the reactor; in the heterogeneous pile it is localized in some form of structural unit.

Each class can be designed in one of three general types: fast, resonance (or intermediate), and thermal. The three types derive their names from the velocity or energy of the neutrons involved in the chain reaction, averaging about 10^5 ev for fast, 10^2 ev for resonance, and 10^{-1} ev for thermal reactors.

Whatever the design of the reactor, those materials in the reactor proper that contain, or immediately surround, the fissionable constituent may be considered as diluents. It is explained in the book "The Science and Engineering of Nuclear Power" (*1*) that "in general, as the atomic weight (A) of the diluent increases, the mean energy at which the neutrons cycle increases." From this and other considerations, it is likely that ceramic materials will be most useful as the moderator in a thermal pile because they are of relatively low atomic weight.

Other important parts of pile construction are the reflector and the shield. The shield must be fabricated of substances that will stop the passage of neutrons and gamma radiations by absorbing or scattering them so as to prevent their direct passage into the surrounding atmosphere. Consequently, the shield is made of elements with high-scattering cross sections and slowing-down power, combined with good absorbers. Hydrogen with a scattering cross section (σ_s) of 21 barns in the resonance region and an energy loss (ξ) of 1 is the best element for scattering and slowing-down. For stationary piles, where weight and size are not a primary consideration, ordinary concrete may be used because the cement contains much water.

It would appear, for the present at least, that the use of ceramic bodies

* This survey was sponsored by, and this report is published with the approval of, the U. S. Atomic Energy Commission.

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may be limited to the reactor core in thermal piles, and particularly in thermal piles operating at 1,000° C or higher. The ceramic would be mixed intimately with the fissionable material to form fuel rods, or used to build the pile's structural, or moderator, portion.

Seven materials are presently under consideration for use as moderators: light and heavy water, beryllium metal, zirconium hydride, beryllium oxide, beryllium carbide, and graphite. Of these, only the last three are "ceramics."

Moderator materials should be effective in slowing down neutrons by virtue of low atomic weight, low-capture cross section, and high-scattering cross section. These terms are explained later. The necessary characteristics of these materials, in addition to their desired nuclear properties, are outlined below.

1. Their resistance to thermal rupture should be high, and, therefore, they should have
   - (*a*) high thermal conductivity
   - (*b*) low thermal dilatation
   - (*c*) low elastic modulus
   - (*d*) high mechanical strength at all service temperatures
2. They should have a high melting (or softening) point
3. They should have high resistance to corrosion by working fluids and low solubility for other elements
4. If they are mixed intimately with fissionable material (as in a fuel rod), it must be possible to
   - (*a*) attain maximum density at the surface
   - (*b*) apply a coating to the mixture to make it impervious both to corrosive elements from without and to passage of fission products from within.

For a more detailed discussion of the service conditions in a heterogeneous reactor core, one may use as a base of reference the greatly simplified sketch of the structural unit shown in Fig. 1. The reflector is not discussed because any material, metal or ceramic, that will satisfactorily withstand the conditions existing in the fuel rod or moderator will certainly be suitable for use in the shell surrounding the reactor core.

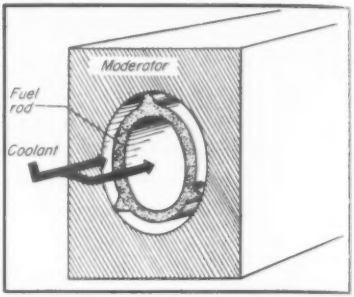

FIG. 1. Cross-section view of a simplified structural unit in a heterogeneous reactor that uses a solid moderator and a gaseous or liquid coolant

The fuel rods, in discrete sections, extend end-to-end through the blocks of moderating material. For example, say they are composed of the fissionable element in an intimate mixture with BeO or Be_2C. Because the heat will be generated within the walls of the rod by the fission process, the rod will be hottest at the mid-portion of the wall. Therefore, a steep temperature gradient will exist between this portion and the wall surfaces in contact with the coolant, placing the surfaces in tension. The tension will be directly proportional to thermal expansion, and inversely proportional to thermal conductivity. Relative thermal stress S may be calculated for a given temperature gradient by means of the equation (*2*):

$$S \sim \frac{E\alpha}{K(1-\mu)} Wh$$

where E = Young's modulus, α = thermal expansion, K = thermal conductivity, μ = Poisson's ratio, W = power production in BTU per sec per cu in., and h = the shape factor.

Today's investigator is fortunate indeed if he has good data on the first three factors and any data at all on the last three, to say nothing of likely temperature gradients, under pile operating conditions. For those interested, there are available theoretical mathematical treatments of heat flow and thermal stresses (*3*, *4*).

It is estimated that the fission of a single nucleus liberates about 198 Mev, equivalent to 3×10^{-14} BTU. This does not seem impressive until one realizes that there may be hundreds of billions, or even trillions, of fissions in a volume of 1 cc every second.

The potential temperature from nuclear fission is expressed in astronomical figures (in the order of $10^9 \times 7.8°$ K) and only by the most careful design and manipulation of control rods and of the coolant would it be possible and practical to prevent instantaneous fusion of the rods and moderator.

Huffman (*5*) states that a pile designed to operate at 1,000 kw could reach 2,700 kw in 0.01 sec if the multiplication constant were to be exceeded by only 0.10. A wide margin of safety between the operating temperature and the softening range of the fuel rod mixture is therefore highly desirable. Although BeO has a melting point of about 2,530° C and Be_2C decomposes above 2,100° C, one could not rely on these values because the fuel rod is a mixture of oxides.

In addition to the uncertainty of maintaining the initial properties of materials inserted in a pile because of thermo-chemical changes, the ceramic engineer must consider, also, that the pile radiations may cause far-reaching changes in physical properties of solids as the nuclear reaction proceeds.

The moderator materials are exposed to high neutron and gamma-ray fluxes. The fuel rods are exposed to these radiations and also to beta particles and fission fragments. As recently emphasized by Weinberg (*5*), "it is to be expected that a solid crystalline material placed in an intense chain reacting pile will suffer damage to its crystals . . . ." It may be assumed that the over-all effect will diminish the resistance of the fuel rod to thermal stresses and this effect is indicated to be greater for mixtures of a ceramic and uranium oxide than for the ceramic alone.

Problems arising from coolant contact. The fuel rod must necessarily be brought into close contact with the coolant to effect the transmission of the heat energy. The following list of possible coolants, used either singly or in combinations, will indicate the interesting engineering problems that must be solved to prevent corrosion and erosion of the fuel rods:

Air (including H_2O vapor)	*Bismuth*	
Helium	*Lead*	
Hydrogen	*Magnesium*	*(molten)*
Nitrogen	*Sodium*	
Carbon monoxide or dioxide	*Potassium*	
Mercury	*Tin*	

In a recirculating system (closed cycle), it appears feasible to adapt an inert gas such as pure helium under pressure. In this case, either graphite, Be_2C, or BeO would be unaffected by the gas. Nevertheless, the danger of diffusing radioactive fission products throughout the circulating system, and into the surrounding atmosphere through even minute leaks, is sufficient to make the use of He far from foolproof.

There are numerous reasons why the other coolants are being considered for specific applications. The use of liquid or of molten metals as coolants would result in appreciable economy of space and fuel for a given energy output. Such coolants would also (*a*) block the escape of neutrons from cooling ports in the reactor; (*b*) serve partially as a reflector; and (*c*) shield against the escape of gamma radiations. Solu-

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tions to the problems arising from the use of metals as coolants in a reactor with a ceramic oxide or carbide moderator have still not reached the point of practical application.

Referring again to Fig. 1, it is seen that the moderator also must be in contact with the coolant. It is more than likely that the maintenance of the moderator, and its protection from reaction with the coolant by means of a coating, will be a simpler problem than that posed by the fuel rod. The moderator will operate somewhat cooler than the rod; it will not be subject to damage from fission fragments; it will undergo less abrupt temperature changes during starting-up and shutting-down operations; it can be of simpler composition (i.e., contain no fissionable material). On the other hand, if the design of the reactor calls for a moderator in comparatively bulky or thick-walled shapes, it may well be that the thermal gradients within the moderator will cause such large thermal stresses as to overcome the advantages. As with the fuel rod, the moderator must have a high resistance to thermal stress; it must have a melting point, or softening range, well above operating temperature, and it probably will need coating.

In the solution of this problem, it is believed that shape design may be ranked equal in importance with composition. The development, and application, of suitable coatings (whether as a barrier for fission products, or as a diaphragm between coolant and ceramic or both) may not be impossible, but it is a challenge to the ceramist.

Calculation of Nuclear Properties (*1*)

Ceramic shapes made from oxides or silicates will, almost without exception, be mixtures rather than pure compounds. The purpose of such mixtures is the same as for metallic alloys. Certain desirable or necessary properties may not be obtainable with the pure compound but can be developed by one or more additions of other materials. The addition of carbon to iron to produce steel is a classic example. In ceramics, the addition of feldspar to kaolin is done to obtain the strength and translucency of china for tableware.

But another factor must be considered when the ceramic is intended for service involving neutron bombardment or subjection to fission fragments. This factor is the nuclear properties of the ceramic mixture. An illustration is the common ceramic glaze which usually contains boron, introduced as borax or as boric acid. So far as ceramic considerations are concerned, there is no objection to boron. Table 1, on pages 8–9, shows however, that this element has a powerful influence on the capture cross section ($N\sigma_c$). A ceramic shape assigned for use as a moderator would be quite unacceptable if it contained even a thousandth of one per cent of boron.

The following properties determine the probable suitability of a composition for pile construction:

Capture (or absorption) cross section (σ_c): The term applies to the fraction of neutrons in a sq cm neutron beam that is removed from the beam, by capture or absorption, in a single nucleus of an element in the path of the beam.

Scattering cross section (σ_s): This term applies to the fraction of neutrons in a sq cm neutron beam that is intercepted, by scattering or reflection, by a single nucleus in the path of the beam.

Energy loss (ξ): Mean logarithmic energy loss per neutron-nucleus collision.

Slowing-down power ($N\sigma_s\xi$): As the symbol implies, this is determined by the fraction of neutrons intercepted, the energy loss per collision, and the number (N) of nuclei per cm³ of a particular element. It is the total logarithmic neutron energy loss by scattering in a centimeter cube of the material.

The number of nuclei per cm^3 is the same as the number of atoms per cm^3, and is a function of the density, or gm/cm^3. When calculating N, it is convenient to express Avogadro's number as 0.602×10^{24} because, when multiplied by the nuclear cross section which is expressed in "barns" (10^{-24} cm^2), the exponents of 10 cancel out.

The values for capture cross section and for scattering cross section are prescribed by the nucleus of an atom, the electrons playing no part in these reactions. Also, the effective cross section will vary with the energy of the neutrons, expressed in electron volts. In this report the cross sections used in the calculations correspond to thermal neutrons with an energy of 0.025 ev.

One method for judging the suitability of additions to a ceramic moderator is based simply on a comparison of the capture cross sections in Table 1 with the cross sections for BeO and for graphite. This method gives values for guidance based on the capture (or absorption) cross section only. It might suffice if the material in question were to be evaluated solely for thermal moderators. Without going into the theoretical considerations, it is sufficient to know that the scattering cross section and the slowing-down power also are important to the nuclear physicist.

The following problem and its solution will illustrate how the three properties may be calculated to obtain values for a first approximation. In this calculation it is assumed that the scattering cross sections are additive in the resonance region. It is well known, however, that, for thermal neutrons, the crystalline structure of a material (specifically the interplaner spacing) is responsible for marked variations in the scatter cross section. These variations are due to interference effects. It must be considered that the crystalline structure of a compound may differ from the structures of the components.

PROBLEM: To calculate (*a*) the overall efficiency, as a structural material for power piles, of a body composed entirely of beryllium oxide (BeO), and (*b*) to compare this with a body composed of 48 moles of BeO, 1 mole of Al_2O_3, and 1 mole of ZrO_2, to 100 parts by weight of which 2 parts by weight is CaO that has been added. The latter body is designated as Body 4811C.

Although BeO has a theoretical density of 3.02 gm/cm^3, a density of 2.8 gm/cm^3 is assumed for the practical manufactured product. Body 4811C can be produced with a density of 3.0 gm/cm^3. Per-cent-by-weight composition of Body 4811C, in terms of the elements, may be calculated as follows:

Determine first the weight % of oxides in the mole composition 48BeO: Al_2O_3: ZrO_2. To this add the 2 weight % of calcia, making 102 "per cent." Recalculate to 100 per cent, which gives

BeO	82.55%
Al_2O_3	6.96%
ZrO_2	8.53%
CaO	1.96%

Then tabulate the constituent elements in each oxide in terms of per-cent-by-weight in the body. These percentage values for each oxide are in the same ratio as the atomic formula weights. See Table 2 on page 10.

The per-cent-by-weight of elements in the all-beryllia body can be calculated in the same way.

The density for each element in gm/cm^3 is next calculated by multiplying the density of the body by the weight % of the element; thus, $29.75 \times 3.0 = 0.89$ gm of Be per cm^3 of Body 4811C.

N, the number of nuclei (atoms) per cm^3 of each element, may now be calculated from the relation:

$$N = \frac{D_e}{W_e} \times 0.602 \times 10^{24}$$

where D_e = density of the element in

NOTE: *Table 1 appears on pages 8–9. Text is continued below Table 2 on page 10.*

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TABLE 1. Physical and Nuclear Properties of Some Ceramic Materials (39).
Four metals and two non-metals are included for comparison purposes.

Material		*Melting point*	*Average coefficient of linear thermal expansion*		*Thermal conductivity*
		°C	°C	($n \times 10^6$)	watts cm^{-1} °C^{-1}
Alumina	Al_2O_3	2,015 ± 15 (40)	25–1,000	8.31 (24)	(870°) 0.04 (41)
Baria	BaO	1,923			
Beryllia	BeO	2,530 ± 30 (43)	25–1,700	10.6 (24)	(200°) 0.8 (600°) 0.42 (1,400°) 0.16
Calcia	CaO	2,570			
Ceria	CeO_2	2,600 (46)	100–500	8.6	
Chromic oxide	Cr_2O_3	2,275 (47)[d]	100–500	7.0	
Cobaltous oxide	CoO	1,935			
Hafnia	HfO_2	2,810			
Lanthana	La_2O_3	2,320			
Magnesia	MgO	2,800	25–1,700	14.7 (24)	(500°) 0.059 (48) (1,000°) 0.036
Niobium oxide	Nb_2O_5	1,520			
Quartz (silica)	SiO_2	1,710	25–500	0.6 ∥ C axis (49) 1.05 ⊥ C axis (49)	(1,000°) 0.02
Thoria	ThO_2	*ca.* 3,000 (50)	25–1,700	10.2 (24)	(150°) 0.095 (400°) 0.050
Titania	TiO_2	1,830	100–500	7.5	
Zirconia	ZrO_2	*ca.* 2,700	70–1,000	7.7 (24)	±0.03
Beryllium carbide	Be_2C	(39B)[b]	38–982	10.8	0.117
Boron carbide	B_4C	2,350	25–800	4.5 (26B)	
Hafnium carbide	HfC	±3,890			
Silicon carbide	SiC	(39B)[e]	20–1,500	4.7 (52)	(?00°) 1.8 (53) (1,100°) 1.0
Tantalum carbide	TaC	3,875 ± 125 (29, 54)[f]	20–2,380	8.2 ± 0.8 (55)	
Titanium carbide	TiC	3,140 ± 90 (56)			
Uranium carbide	UC_2	*ca.* 2,260			
Wolfram carbide	WC	2,870 ± 50 (29)	20–1,930	5.2–7.3 (55)	
Zirconium carbide	ZrC	3,530 ± 125 (29)			
Aluminum (57)	Al	660.2	20–600	29.4 (58)	
Beryllium (57)	Be	1,284	20–200	12.4	0.16
Boron	B	2,300	20–750	8.3	
Cadmium	Cd	320.9			
Graphite	Cg	3,980	25–1,800	3.34 (52)	0.6–3.5 (59)
Zirconium (57)	Zr	1,750 ± 700	rm. temp.	5–6.3	

[a] The density values are those used in calculating N. σ_c and σ_s correspond to a neutron energy of 0.025 ev.
[b] 18,500 psi in tension at 1,200° C.
[c] Value for impervious specimens containing 84.2 weight per cent of BeO is *ca.* 14,000 psi at 982° C. Ref. 42 gives 2,100 psi in tension at 1140° C.
[d] Cr_2O_3 volatilizes rapidly above 2000° C.
[e] Sintered specimens.
[f] Brick specimen containing about 91 weight per cent of MgO.

(Numbers in parentheses refer

Elastic modulus room temp.	*Modulus of rupture (in bending, unless otherwise noted) rm. temp.*	*Hardness: K—Knoop M—Mohs*	*Total nuclei per cm³*	*Nuclear Properties (1, 6)*			*Density*[a]
				Cross section in barns		*Slowing-down power*	
				$N\sigma_c$	$N\sigma_s$	$N\sigma_s\xi$	
($psi \times 10^{-6}$)	*psi*		($N \times 10^{-24}$)	(cm^{-1})	(cm^{-1})		(*gm/cc*)
55 (*42*)	46,800 (*42*)[b]	*K*: 2,150 (*27*) *M*: 9	0.112	0.010	0.342	0.038	3.8
			0.045	0.028	0.272	0.014	5.72
45 (*42*)	20,600 (*44*)[c]	*K*: 1,250 (*45*) *M*: 8	0.1335	0.0007	0.68	0.114	2.8
		M: 4.5	0.073	0.182	0.241	0.022	3.4
		M: 6	0.0756	0.016	0.333	0.029	7.2
			0.1032	0.103	0.419	0.036	5.2
			0.1038	1.868	0.473	0.034	6.47
			0.083	2.94	0.505	0.030	9.68
			0.060	0.192	0.459	0.061	6.5
		K: 370[e]					
12.4 (*43*)[e]	2,800[f]	*N*: 6	0.1104	0.0033	0.419	0.042	3.6
			0.0709	0.024	0.309	0.027	4.47
		K: 820 (*27*) *M*: 6–7	0.0797	0.0064	0.279	0.029	2.65
21 (*43*)[e]	17,900 (*42*)[e]		0.066	0.132	~0.400	0.023	9.69
		M: 6 ± 0.5	0.0963	0.167	0.456	0.039	4.26
36	(*51*)	*K*: 1,160 *M*: 6.5	0.084	~0.01	0.460	0.036	5.72
	ca. 25,000 (*45*)	*K*: 2,410 (*45*) *M*: 9+	0.147	0.0012	0.793	0.154	2.4
65	44,000 (*26B*)	*K*: 2,750 (27) *M*: 9+	0.138	80.8	0.553	0.306	2.5
			0.077	4.05	0.570	0.032	12.2
		K: 2,500 (*45*) *M*: 9+	0.0952	0.0116	0.338	0.042	3.17
41	ca 3,500[f]	*K*: 2,000 *M*: 9+	0.0904	0.904	0.434	0.036	14.48
46		*K*: 2,470 *M*: 9+	0.0852	0.222	0.461	0.041	4.25
			0.081	~0.054	0.480	0.041	11.78
74	250,000 (*26B*)	*K*: 1,880 *M*: 9+	0.0962	0.866	0.471	0.036	15.7
	6,875–16,300[f]	*M*: 8–9 (*55C*)[k]	0.080	~0.016	0.520	0.036	6.9
			0.0536	0.011	0.072	0.005	2.7
42	17,000–27,000[f]	Brinnell 97 to 114	0.120	0.0012	0.732	0.146	1.8
		M: 9.3	0.139	101.4	0.528	0.09	2.3
			0.0461	133.6	0.300	0.012	8.64
1.4–2.2	2,800–5,000	Scleroscope 25 to 45 (*60*)[m]	0.0812	0.0004	0.390	0.058	1.62
11.3–14.5	36,000–80,000[f]		0.0422	~0.016	0.346	0.007	6.4

[a] An approximate value obtained at the National Bureau of Standards on a polished face of a water-clear crystal cut at 123° to the cleavage.
[b] Decomposes above 2,100° C.
[c] Decomposes above 2,200° C.
[f] Value for stress in tension.
[k] Brinnell: annealed, 17; cold-rolled, 27.
[l] There is some evidence that TaC decomposes before melting.
[m] Vickers hardness for unworked metal, 82 kg/mm² using 10 kg load.

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TABLE 2

Percentage by Weight in the Body of Constituents in Four Oxides

Body 4811C	*Oxide*	Be	Al	Zr	Ca	O
BeO	82.55%	29.75%	.....	.....	.....	52.80%
Al_2O_3	6.96	.....	3.69%	.....	.....	3.27
ZrO_2	8.53	.....	.....	6.31%	.....	2.22
CaO	1.96	.....	.....	.....	1.40%	0.56
		29.75	3.69	6.31	1.40	58.85

gm/cm³, W_a = atomic weight of the element, and 0.602×10^{24} = Avogadro's number.

The calculation of the value N may be expedited by calculating the number of nuclei for one element and determining the others by simple proportion of the atoms.

The calculation of the pertinent nuclear properties [the nuclear values may be obtained from the literature (*1, 6*)] now consists simply of multiplying the respective nuclear values per nucleus by N and obtaining a summation. See Table 3.

With the values from Table 3, it will be found convenient to arrange the nuclear properties as shown in Table 4.

The results show BeO to have an appreciably lower capture cross section than Body 4811C, but a slightly higher scattering cross section and slowing-down power. The relative importance of the properties will be governed by the type of power pile, and the location in which the material is to be used.

Discussion of Materials

The following discussion supplements the information presented in Table 1.

Alumina. This compound is the most available in nearly pure form of those oxides melting near, or above, 2000° C. Shapes of Al_2O_3 may be formed by casting, extrusion, or pressing, and matured by heating to an impervious structure resembling porcelain in appearance. Shapes so matured are stable in air, resistant to attack by both acids and bases, and are claimed to be impervious to gases at 1700° C and resistant to reducing gases such as H_2 and CO. In addition, sodium

TABLE 3

Summation of Nuclear Properties

Element	*Composition* wt %	*Composition* gm/cm³	N*	σ_c barns	σ_s barns	ξ
Compound BeO						
Be	35.8	1.00	0.067	0.009	6.1	0.2
O	64.2	1.80	0.067	< 0.001	4.1	0.12
Body 4811C						
Be	29.75	0.89	0.060	0.009	6.1	0.2
Al	3.69	0.11	0.0025	0.23	1.5	0.07
Zr	6.31	0.19	0.0012	0.4	14.0	0.02
Ca	1.40	0.04	0.0006	0.43	9.5	0.05
O	58.85	1.77	0.066	< 0.001	4.1	0.12

* Nuclei per cm³, to be multiplied by 10^{24}.

carbonate, caustic soda, and even sodium peroxide may be fused in pure alumina vessels with little attack (*7*).

It would appear (*8, 9*) that bodies of pure alumina (over 99.5% of Al_2O_3) must be used to develop its full potentialities as regards mechanical strength. However, its high elastic modulus and intermediate thermal conductivity (compared with other ceramic oxides) militate against its resistance to thermal shock. Al_2O_3 is not at present interesting as a moderator, principally because of the relatively large capture cross section, but it might be considered in other applications for nuclear reactors.

Baria. For use in a reactor, barium oxide has ample resistance to fusion but its poisonous nature, lack of stability in air, and unfavorable nuclear properties explain the dearth of information on its properties.

Beryllia. This oxide is unique because of its high thermal conductivity (possibly 15 to 20 times greater than that of alumina) and its high electrical resistivity higher than the resistivity of alumina or magnesia (*10*)].

The following values were obtained for the pure oxide: the crushing strength at 1200° C was 28,000 psi, the modulus of elasticity was 20×10^6 psi and, at 1130° C, the tensile strength was about 2,000 psi (*11*). The high thermal conductivity of BeO, favoring high resistance to thermal shock, and the relatively low capture cross section, make it one of the most favored of the ceramic materials for use as a moderator. However, beryllia is attacked by water vapor at elevated temperatures (*12*) which may necessitate its protection from moisture-containing coolants in a reactor.

Recently, attention has been called to the possible toxic effects of beryllium compounds (*13*). The use of efficient ventilation and the most careful personal hygiene is indicated.

Calcia. This very refractory oxide is one of the most plentiful in the earth's crust but may be placed in the same category as baria so far as its present usefulness in reactors is concerned.

Ceria. Little is known about the properties of ceria. In refractoriness, it compares favorably with calcia; in addition, it is stable in air. In the pure form, ceria is rather expensive, small lots costing about $20 per pound. The electrical resistivity values of ThO_2-CeO_2 mixtures at 800° and 1200° C in atmospheres of O_2, N_2, and H_2 have been reported (*14*). CeO_2 is said to reduce, even in oxidizing atmosphere, to Ce_2O_3 which fuses at 1690° C (*15*); however, a specimen heated to 2000° C in air at the National Bureau of Standards (*16*) did not fuse and showed no evidence of reduction.

Oxides of chromium, cobalt, and niobium (columbium). There is no information available regarding the fabrication of the pure oxides of these elements into useful articles, and the only reason for their inclusion in Table 1 is their refractoriness. Con-

TABLE 4

Cross Section and Slowing-down Powe

Element	$N\sigma_c$ (cm^{-1})	$N\sigma_s$ (cm^{-1})	$N\sigma_s\xi$ (cm^{-1})
Compound BeO			
Be	0.00060	0.409	0.0818
O	0.00006	0.274	0.0330
	0.00066	0.683	0.1148
Body 4811C			
Be	0.00054	0.3660	0.07320
Al	0.00057	0.0037	0.00026
Zr	0.00048	0.0168	0.00336
Ca	0.00026	0.0057	0.00028
O	0.00006	0.2710	0.03250
	0.00191	0.6632	0.10960

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ventional chrome refractories contain from 35% to 65% of chromic oxide and are noted for their high resistance to both acid and basic slags; they are sensitive to reducing conditions and thermal shock (*17*).

Oxides of hafnium and lanthanum. Neither of these oxides is readily available, hafnia, in particular, being extremely scarce because of its difficult separation from zirconia, or zirconium, in which it occurs as a highly undesirable impurity because of its high capture cross section. Lanthana may be purchased in small lots at about $20 a pound, but it absorbs moisture so greedily as to be quite unstable in air. The refractoriness of these oxides and the high scattering cross section of lanthana are the only reasons for their inclusion in Table 1.

Magnesia. Magnesium oxide occurs in nature as the mineral periclase, but is rare. Fused and recrystallized magnesia (periclase) of 96% to 97% purity is readily available, and pulverized material of better than 99% purity has been obtained commercially at less than $1 per pound. Ryschkewitsch (*7*) states that MgO is an ideal refractory for withstanding the attack of molten PbO, and that impervious as well as porous articles can be fabricated of MgO. He states also that, in oxidizing atmospheres, MgO is no more volatile than other highly refractory oxides. In vacuum, however, it is known to volatilize at temperatures as low as 1600° C (*18*).

Whenever MgO was the major constituent in binary- or ternary-oxide mixtures investigated at the National Bureau of Standards (*19*), the resultant bodies were coarse-grained and comparatively weak in mechanical strength. The high thermal expansion and low thermal conductivity indicate a relatively poor resistance to thermal shock.

Magnesia also, in common with calcia, will combine with moisture and especially so at elevated temperatures. It is known that the presence of certain impurities in periclase refractories (*17*) will increase resistance to water vapor, and the problem of "stabilizing" magnesia and calcia is being investigated (*20*).

The rather low capture cross section of MgO is interesting but, in the present state of the art, it is doubtful if suitable moderators of pure, or nearly pure, MgO could be fabricated.

Silica. Silica is a material of many forms (*21*). Crystal phases are usually referred to as alpha and beta quartz, alpha and $beta_1$ and $beta_2$ tridymite, alpha and beta cristobalite, and glass. With the exception of the $beta_1$–$beta_2$ inversion, changes from one crystalline form to another are accompanied by very abrupt changes in volume. Expressed as change in length, they range from about 0.15% for the alpha–$beta_1$ tridymite inversion, to 1.08% for the alpha–beta cristobalite inversion. Because of these abrupt volume changes, and poor thermal conductivity, any shape containing an appreciable quantity of crystalline SiO_2 will have poor resistance to thermal shock.

The glass form of silica (often referred to as "fused quartz") has a very low thermal dilatation. The average coefficient of linear expansion between 20° and 1000° C was found to be 0.48×10^{-6} per °C (*22*). Valuable for its excellent resistance to thermal shock through moderate temperature ranges, it loses strength and devitrifies rapidly above about 1100° C.

All in all, silica and the silicates show no promise for use in nuclear reactors.

Thoria. This material has the highest known melting point of the ceramic oxides and has a very low vapor pressure at elevated temperatures. Thoria is

reported (*11*) to have strength in compression ranging from 213,000 psi at room temperature to 1,400 psi at 1500° C. The same source reports the modulus of elasticity ranging from 21 × 10^6 psi at room temperature to 13 × 10^6 at 1200° C. Investigations at the National Bureau of Standards (*19*) indicate that binary- or ternary-oxide porcelains containing more than 70 to 80 weight % of ThO_2 will not, practically speaking, mature to an impervious condition.

Shapes of thoria may be fabricated by procedures similar to those used for other non-plastic ceramics such as BeO, ZrO_2, or Al_2O_3 (*23*), and it is normally available commercially. Chemically, ThO_2 may be considered as basic and it is radioactive. In closed containers, sufficient radioactive gas may be produced during prolonged storage to constitute a health hazard. There are at present no reasons to suppose that shapes of ThO_2 will be used in constructing reactors.

Titania. The potentialities of this oxide, *per se*, as a reactor component are probably nil because of its high capture cross section and the difficulty of maturing it to a dense, strong, shape without at least partial reduction to a lower form of oxide.

Zirconia. The oxide of zirconium is readily available in pulverized form containing about 97 weight % of ZrO_2, and can also be obtained commercially in purity of 99% of ZrO_2. Its chemical inertness to both acids and bases, and its high melting point, make it an interesting material for commercial refractories. Unfortunately, the large and abrupt volume changes accompanying the monoclinic $\rightleftharpoons$ tetragonal inversions cause extreme sensitivity to thermal shock, and the purer the material, the greater are these volume changes. The so-called "stabilized zirconia" (containing small additions of either MgO or CaO) is much more resistant to thermal shock but still not of high resistance. This is so because even the stabilized material has a rather high thermal dilatation [average coefficient about 11 × 10^6 (*24*)], the thermal conductivity is low, and the elastic modulus is high.

The oxide of hafnium very closely resembles the oxide of zirconium chemically. Consequently, the two are difficult to separate and investigations are now in progress on that problem. This separation must be made before either the metal zirconium, or its oxide, can be of interest to nuclear engineers, as is evident from the high capture cross section of hafnia (Table 1).

Beryllium carbide. Although this carbide is one of the three most highly favored "ceramic" materials for reactor construction (the others being beryllia and graphite), very little is known about its properties. The dearth of information may be charged mainly to the difficulties of fabricating shapes, and to the lack of stability in air containing water vapor at room as well as elevated temperatures. Regarding the stability of Be_2C in air, the following conclusions are taken from a NEPA survey (*25*):

Compared to graphite, pressed Be_2C appears fairly stable in dry air. Rod samples of graphite were completely oxidized within 3 to 4 hours at 1000° C, whereas pressed Be_2C will withstand temperatures up to 1593° C for several weeks in dry air with a resultant oxidation to a depth of only a few thousandths of an inch. In moist air, disintegration of pressed Be_2C was complete at room temperature in ten weeks and, at 204° C, disintegration was complete within 4 days. At temperatures above the thermal decomposition temperature (about 200 to 300° C) of $Be(OH)_2$, however, the reaction produces a protective coating of BeO or Be suboxides. The density of the Be_2C is one of the determining

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factors. For example, at about 1250° C and a moisture content expressed as 28 mm H_2O pressure, a specimen of 1.998 gm/cc density gained in weight 0.0823 gm/cm² of surface during one week of exposure. Under the same conditions, the weight gain for a specimen of density 2.323 gm/cc was 0.0004 gm/cm² of surface.

From the data available, it seems conclusive that the usefulness of Be_2C in service conditions involving exposure to water vapor will be contingent upon the development of a suitable coating.

Boron carbide. This material was first described by Ridgway (*26*). The production process is similar to that of silicon carbide and the reaction temperature is 2500° to 2600° C. B_4C has a very low vapor pressure at high temperatures and does not distill. The work of Ridgway indicates boron carbide to be harder than silicon carbide, and this was later substantiated by Thibault and Nyquist (*27*). Molded boron carbide showed a compressive strength of 255,000 psi and an electrical resistivity ranging from 0.445 ohms-cm at 20° C to 0.023 ohms-cm at 500° C.

For mechanical strength and hardness, B_4C is outstanding. However, its rate of oxidation is about ten times that of silicon carbide. Its capture cross section also is unfavorable to its use in a reactor except as a control.

Hafnium carbide. The principal claim to notoriety of hafnium may well be its nuisance value as an impurity in zirconium compounds. The fabrication methods and properties of hafnium compounds may be expected to parallel closely those of the similar compounds of zirconium. Its foreseeable availability and usefulness in nuclear reactors are very low.

Silicon carbide. The natural carbide has the mineral name *moissanite*. The artificial electric-furnace product was first made in 1891 by E. G. Acheson and has since become known and used throughout the world as an abrasive and as a refractory. The relatively high thermal conductivity and low thermal expansion favor good resistance to thermal shock. Very little has been published on the properties of SiC *per se*, but the bonded and the recrystallized products have been the subject of numerous reports (*28*). In air, the oxidation probably will be negligible at 1000° C and rapid at 1500° C. Published information on this point is confusing and often contradictory.

SiC is subject to large-scale production methods and is readily available. Shapes containing a high percentage of SiC also may be fabricated by any one of several well-established methods. However, its potentialities in power piles, except as a possible coating material, are believed to be poor.

Tantalum carbide. This carbide has an extremely high melting point, approximating the melting points of hafnium carbide and of graphite (*29*). It is essentially metallic in nature and a good conductor of electricity. No values were found for thermal conductivity but this, probably, is also high. Tantalum metal in fabricated forms may be purchased at from $100 to $150 per pound. From this, and from its occurrence in the earth's crust (estimated less than 0.0015%), it may be deduced that, in the present state of the art, tantalum carbide also would have rather limited availability. The nuclear properties (Table 1) are not such as to make the material of interest in reactor construction at this time.

Titanium carbide. Published information on this carbide is extremely limited, and it is included in the report only because of its high melting point and hardness. Regarding its stability

in air at elevated temperatures, Becker (*30*) states that it is among the most resistant of the metal carbides, but he presents no quantitative data. From nuclear considerations, there would be no objection to its limited use.

Uranium carbide. As is indicated in Table 1, little is known about the properties of UC_2. It could be used as a solid fuel providing its fabrication can be accomplished commercially and providing its resistance to corrosion, resistance to thermal stresses, and other essential prerequisites are favorable (*1*). Recent investigations have proved the existence also of the monocarbide UC, which is thermodynamically stable at room temperature while the dicarbide is stable only at high temperatures. It is reported that U_2C_3 exists above 2000° C.

Wolfram (tungsten) carbide. The data in Table 1 are confined to the monocarbide although the wolfram carbides include also W_2C and cobalt-cemented WC. The pure carbides are characterized by extreme hardness, but are too brittle for industrial application. The addition of cobalt binder increases the toughness markedly, and certain of the cemented carbides are reported to have the highest elastic modulus of any known material. Addition of cobalt, however, reduces the extreme high-temperature usefulness because of the melting point of the binder (~1480° C).

Zirconium carbide. The low capture cross section and high melting point of ZrC, combined with apparent good mechanical strength (at least at room temperature), favor its use as a moderator material. This presupposes the absence of hafnium.

Nitrides of beryllium, boron, tantalum and zirconium. These four nitrides have high melting points. Information on other physical properties is so meager that it would be impossible to evaluate their potential uses. Boron nitride sublimes before melting at atmospheric pressure (*31*). If it should find use in a reactor, it would most likely be as a control. Tantalum nitride is reported to have a high melting point. Nothing else was learned about its physical properties, and the cross sections are not in the range of interest. Zirconium nitride is characterized by its high melting point. It combines readily with nitrogen. In addition to the mononitride, Zr_3N_2, Zr_2N_3, Zr_3N_4, and Zr_3N_8 have been reported.

Hydrides of tantalum, thorium, uranium and zirconium. An article by Gibb (*32*), together with its 44-item bibliography, is a good reference source on these hydrides, but contains no data of help to the nuclear engineer.

Aluminum. The "Metals Handbook" (*33*) summarizes the present knowledge on the physical properties of aluminum. Next to silica, its oxide is the most abundant in the earth's crust. It is of interest to pile designers for use as a structural material in low-temperature reactors.

Beryllium. Recent estimates (*34*) indicate that Be constitutes about 0.0005% of the earth's crust. It is, therefore, one of the rarer elements. The "Metals Handbook" (*33*) summarizes information on physical properties.

Boron and cadmium. These elements are among the most valuable materials for control purposes because of their large capture cross section. Boron is a non-metal and occurs to the extent of about 0.001% of the earth's crust. Very difficult to prepare in the elemental form, it is seldom made as such except for laboratory use (*35*).

Cadmium is considered the rarest

of the common metals, but its usefulness is limited by its low melting point.

Graphite. This non-metal has of late been considered a "ceramic." Its extreme refractoriness, low capture cross section, and high resistance to thermal stresses brought it to the attention of the nuclear physicists early in the development of pile technology. Many tons of high purity graphite were used by the Manhattan Project. The vapor pressure ranges from 0.04 mm at 2375° C to 760 mm at 4190° C (*36*). Thermal expansion varies with crystal direction, the value in Table 1 being an average for a specimen of presumed random orientation. Thermal conductivity also is anisotropic and seemingly, in formed shapes, is greater in the direction in which the long axes of the graphite particles tend to be oriented. The electrical resistivity likewise varies with crystal direction.

Zirconium. Comprising at least 0.01% of the earth's crust, this element may be considered as abundant. Its preparation as the pure metal, however, is still expensive. The metal is very stable chemically, and resists both acidic and alkaline solutions. It is pyrophoric, and combines with all but the rare gases at high temperatures. It is used as a "getter" metal.

Values for the coefficient of linear thermal expansion given in Table 1 (*35*) are average. One report (*37*) gives the coefficient 2.5×10^{-6} parallel to, and 14.3×10^{-6} perpendicular to, the hexagonal axis. It is probable that much of the currently available information on both physical and nuclear properties (*38*) will be modified when further tests are made on the hafnium-free, high-purity, metal.

Conclusion

The use of ceramics, or metal-ceramic combinations, for shields, moderators, fuel rods, and coatings in thermal piles operating at temperatures in excess of about 1000° C offers the only practical alternative to the use of metals in lower-temperature piles.

The survey of ceramic potentialities, in the light of present knowledge as presented in this report, shows that many difficult and complex problems remain to be solved. These problems range from the mass fabrication of parts, and the application of continuous protective coatings, to the development of additional fundamental information on the ceramic materials themselves and phase relations of the systems involved.

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272 MDR-11 Boron carbide
310 TID Zirconium
408 EMR-22 Silicon carbide
427 EMR-26 Silicon oxide
437 IHR-E12 Zirconia
463 IHR-E14 Magnesia
465 EMR-27 Tungsten carbide
531 EMR-29 Tantalum carbide
719 EMR-31 Beryllium carbide
875 EMR-42 Thoria
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Response of G-M Counters and Photographic Emulsions to High-Energy Photons

Emission of secondary electrons produced by absorption of photons (0.5–10 Mev) in matter is calculated. Results are in good agreement with experimental counter efficiencies. A qualitative explanation of the intensifying action of low and high atomic number materials surrounding a film is given

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CHARGED PARTICLES, such as electrons, protons or α-particles, form ion pairs along their path through matter. Therefore they will be registered if they enter the sensitive part of a measuring device. Nonionizing electromagnetic radiation such as X- and γ-rays, however, can pass through an ionization chamber, a Geiger-Müller counter or a photographic emulsion without any noticeable reaction. The high-energy photon first has to produce an electron; then this electron must enter the detector to register the primary photon indirectly. The probability for detection of an X- or γ-ray will therefore depend on the secondary-electron production rate and the range of these electrons in various materials.

It is not the purpose of this paper to give a complete discussion of the different radiation detectors or to evaluate their efficiency for photons of various energies. Rather, it is intended to show how the response of Geiger-Müller counters and photographic emulsions to high-energy photons depends in a similar way on the secondary electron emission of the surrounding media. The efficiency of Geiger-Müller counters can be calculated, and experimental data are available from a recent investigation with γ-ray energies up to 2.62 Mev. On the other hand, very little is known at present about the response of photographic emulsions to photons with energies of more than about 200 kev.

Data will be given here showing the relative variation of the photographic response of films exposed to X-radiation up to a maximum photon energy of 10 Mev. The photographic emulsion was surrounded by low and high atomic number materials, corresponding to the different cathode materials of Geiger-Müller counters. The intensifying action of high-Z materials for photons with energies between 0.5 and 10 Mev will be calculated and compared with the experimental results. A method for the calibration of the film response will be indicated.

The absorption coefficients of high-energy photons (0.1–10 Mev) in air, aluminum and lead have been given recently as a function of the photon energy (*1*, *2*). From these data, the production rate of secondary electrons by the photoelectric effect, the Compton scattering process, and pair production

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can be derived. The amount of energy given to the electron by each of the three processes can be calculated. Finally, if the corresponding ranges of the secondary electrons are also known, the electron emission of a material under high-energy photon irradiation can be calculated.

Energy of Secondary Electrons

If $E_{h\nu}$ is the energy (in Mev) of the absorbed photon, the energies of the secondary electrons can be expressed in the following manner for the three processes.

Photoelectron. The energy of the photoelectron will be

$$E = E_{h\nu} - W_K \qquad (1)$$

where W_K is the binding energy of the electron to its atomic shell. That means high-energy photons can transfer practically all their energy to an electron.

Compton electron. Depending on the angle between the incident photon and Compton electron, the latter will receive all energies up to a maximum energy of

$$E_{max} = E_{h\nu} \frac{1}{1 + \frac{0.511}{2\,E_{h\nu}}} \qquad (2)$$

Since $\sigma_a + \sigma_s = \sigma$ represents the probability for the occurrence of a Compton interaction (total Compton coefficient), and σ_a is a measure of that fraction of the initial photon energy converted to kinetic energy of the Compton electron, σ_s or $(\sigma - \sigma_a)$ is that part of the remaining energy which is emitted as a scattered photon (*2*). Therefore the average energy of the Compton electrons is given by

$$E_{av} = E_{h\nu} \frac{\sigma_a}{\sigma_a + \sigma_s} \qquad (3)$$

Electron-positron pairs. Photons with $E_{h\nu} > 1.02$ Mev can produce electron-positron pairs. Of the photon energy, 1.02 Mev is used for the materialization of the photon. On the average the excess energy will be equally shared

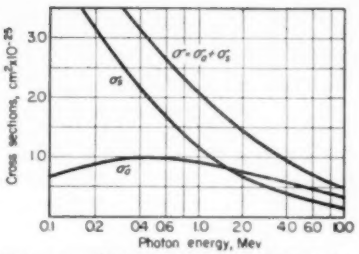

FIG. 1. Klein-Nishina coefficient for Compton scattering per electron of material as a function of the photon energy

by the electron and positron. Therefore each of the two particles will have an energy

$$E_{e^-,e^+} = \frac{1}{2}(E_{h\nu} - 1.02) \qquad (4)$$

Production Rate

Concerning the production rate of these secondary electrons, the probability for the photo-effect is very small for photons with energies greater than 0.5 Mev. Only in materials of high atomic number ($Z > 50$), is the photo-effect appreciable for high-energy photons (*1*, *2*).

Since the Compton interaction is a collision of the photon with an atomic electron, its probability is proportional to the number of electrons per gram of material. Therefore σ, σ_a and σ_s can be presented as a function of the photon energy, independent of the density and the atomic number Z of the material. Their values are plotted in Fig. 1 per electron of absorber for photon energies up to 10 Mev. The number of electrons, n, per gram is given as

$$\frac{n}{\text{gm}} = \frac{NZ}{A} \qquad (5)$$

where N is Avogadro's number and A the atomic weight of the material. From Fig. 1 it follows that the initial energy of photons with energies greater than 1 Mev is, on the average, nearly equally distributed between the sec-

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ondary electron and the scattered photon. Therefore the average energy of the Compton electrons is $E_{av} \sim \frac{1}{2} E_h$, for the high-energy region.

Finally, the pair production process becomes important only if high-energy photons pass through matter of high atomic number. For a 10-Mev photon the probability of pair production in lead is more than three times that of the Compton process, while in aluminum this ratio is only one half.

Range in Matter

The range, or better, the practical maximum range of monochromatic electrons in various materials, has not been investigated very thoroughly. Most measurements have been made with aluminum as absorbers (*3*, *4*, *5*), and very few data are available for heavy materials (*4*, *6*). Electrons of intermediate energy passing through matter lose their energy mainly by inelastic collisions with atomic electrons. Positive and negative ions, formed along the electron path, are the result of this interaction. High-energy electrons expend part of their energy in the production of radiation, called Bremsstrahlung. The ratio between radiation and ionization losses is proportional to the energy of the electron and the atomic number of the absorber. Electrons with energies of 10 Mev passing through lead spend their energy equally by radiation and ionization. However, for electrons of lower energy and absorbers of low atomic number material, the radiation loss is negligible.

Except for a small correction depending logarithmically on Z, the ionization losses are proportional to the number of atomic electrons, n, per cubic centimeter. This number follows from Eq. 5 as

$$\frac{n}{\text{cm}^3} = \frac{NZ}{A}\rho \qquad (6)$$

where ρ is the density of the material. Thus the fraction of energy dE lost by ionization along the path of length ds is, for any particular velocity

$$dE \sim N\left(\frac{Z}{A}\right)\rho\, ds \qquad (7)$$

If the thickness of the penetrated material is expressed in gm/cm², that is, ρds, it follows from Eq. 7 that the ionization energy loss of moving electrons is proportional to (Z/A).

With the exception of hydrogen, the ratio (Z/A) of atomic number to atomic weight is nearly constant for all elements, decreasing from $Z/A = 0.5$ for He, C, O, Al, and Ca to $Z/A = 0.4$ for lead. Therefore, the relation given by various authors (*3*, *4*, *5*), based on measurements made with aluminum as absorbing material, is applicable to all kinds of absorbers. With the exception of the absorption of high-energy electrons in high-Z material, the slight decrease in Z/A and in the logarithmic correction terms with increasing Z is almost compensated by the increased scattering of the electrons in the heavy elements. Experimental data (*6*) indicate a reduced practical maximum mass range in lead. The results of these measurements, however, depend very much on the geometry of the experimental set-up.

Efficiency of G-M Counters

The photon efficiency of a Geiger-Müller counter is the ratio of the number of secondary electrons entering the counter volume per photon passing through the counter. Since high-energy photons are mainly absorbed in the walls of the counter, only those electrons can reach the counter volume which are produced in a layer of the wall equal to the range of the electrons. Based on the preceding discussion concerning the production of secondary electrons as a function of the photon energy and some simplifying assumptions, the counter efficiency ϵ can be calculated (*7*, *8*) as

$$\epsilon \sim (\tau R_\tau + \sigma R_\sigma + 2\kappa R_\kappa) \qquad (8$$

where τ, σ and κ represent the probabilities for the γ-ray absorption by photoeffect, Compton scattering process and pair production, all given per gm/cm² of the absorber, and R_τ, R_σ, and R_κ are the corresponding ranges (in gm/cm²) of the emitted electrons. Since Compton electrons and electron pairs produced by a monochromatic γ-radiation both have a continuous energy distribution, the electron ranges have to be taken for their average energies, as given by Eqs. 3 and 4. In the simple expression for the counter efficiency (Eq. 8), the diminution of the photon beam passing through the counter has been neglected. Furthermore, no geometrical corrections are made for the angular distribution of the secondary electrons relative to the direction of the photon beam.

The variation of the counter efficiency with the energy of the γ-rays given by Eq. 8 is in good agreement with the experimental data obtained by Bradt et al. (7). The counter efficiency had been determined with the γ-rays of various radioisotopes for counters with aluminum and lead walls.

When Eq. 8 was employed as suggested by von Droste (8), the agreement between the calculated and experimental values was not as good. Instead of the range for the average energy of the Compton electron spectrum (Eq. 3), the range for its maximum energy (Eq. 2) had been used. The resulting value was then multiplied by a constant factor of 0.6, determined experimentally by von Droste.

It is quite obvious that this approximation can be a good one for only a small energy interval. The shape of the Compton spectrum changes with its maximum energy, and the ratio of the latter to the average energy is not constant. Furthermore, the electron range–energy relation also is not linear.

The relative efficiencies of counters with walls of different materials can be

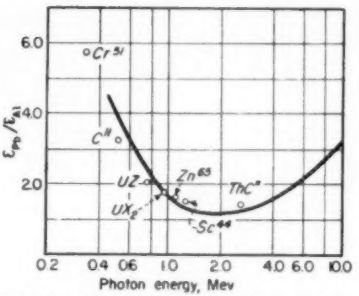

FIG. 2. Calculated ratio of the secondary-electron emission of lead and a low atomic number material irradiated by 0.5–10 Mev photons. Experimental values for the relative efficiency of lead and aluminum counters for six γ-ray energies (7)

derived from Eq. 8. For elements of light and intermediate atomic number, and γ-ray energies between 0.5 and 3 Mev, the efficiency of two counters will be the same, since all secondary electrons are produced by Compton scattering. This is in accordance with the experimental results obtained by Bradt, *et al.* (7) by comparing the efficiencies of brass and aluminum counters with the annihilation radiation of C^{11} (0.511 Mev) and the ThC″ γ-rays (2.62 Mev). In this energy interval the ratio of the efficiencies of brass (copper) and aluminum counters is unity.

In heavy materials such as lead, photoelectrons are produced by γ-rays with energies up to several Mev, and a considerable number of electron pairs originate from γ-rays with energies greater than 1.5 Mev. Therefore counters with a lead cathode show a high efficiency in the low- and high-energy region (7).

The ratio of the electron emission of lead and a material with an atomic number of $Z = 7$ has been calculated as a function of the photon energy by means of Eq. 8 and plotted in Fig. 2. The atomic number of air and tissue, $Z = 7$, was chosen for reasons to be dis-

cussed later. However, since practically all secondary electrons in light material are Compton electrons, the values of Fig. 2 hold just as well for aluminum. The experimental values obtained by Bradt *et al.* with γ-rays of five different isotopes, comparing lead and aluminum counters, check very well with these calculated data (Fig. 2). This shows that the electron emission of a material irradiated by high-energy photons is well represented by the simple expression that has been given in Eq. 8.

Interaction with Photographic Material

Under various experimental conditions, the photographic plate is the most convenient radiation detector. Since it requires very little space it does not change the radiation field significantly and thus allows the registration of the field's spatial distribution. Furthermore, it integrates the radiation incident over a certain length of time and is therefore particularly useful for the determination of the radiation dose delivered by X- or γ-rays (*9*, *10*). But the calibration of the film response as a function of the photon energy cannot be accomplished in such a simple way as the calculation of the efficiency of a G-M counter.

Several papers have been published recently (*11–14*) detailing investigations of the mechanism of the photographic action of photons of various energies. The processes involved are the same as those already discussed; production of secondary electrons in the photographic material varies with its density and atomic composition. The photographic sensitivity, however, depends not only on the number of electrons produced, but also on their energy, which determines their range and therefore the number of ion pairs produced in the absorber. Therefore, the spectral film sensitivity (in reciprocal r units) is a rather complicated function, especially in the low-energy region (*12*). For example, the mass absorption coefficient of silver bromide is high in comparison to air for photon energies below 100 kev. However, 25.5 kev and 13.5 kev of the photon energy are spent for the photoelectron production (Eq. 1), corresponding to the binding energy of the *K*-electrons of Ag and Br respectively. Therefore a photoelectron originating from a 78-kev photon absorbed in AgBr will have, on the average, only an energy of 55 kev, and a mean range in silver bromide of about 10 μ. This corresponds to an effective range of eight grains using the Ansco Non-screen X-ray emulsion (*13*). Since the ion density of low-energy electron varies considerably along its path, all grains probably will not be affected equally. The spectral film sensitivity is therefore also a function of the selected emulsion and developing process. All these facts prevent the successful calibration of X-ray film for low-energy X-rays. A small change of the amount of X-ray filtration, affecting the spectral X-ray distribution only slightly, might change the photographic action of the X-rays considerably.

At higher energies (above 300 kev) the photoelectric absorption is no longer predominant. The production of Compton recoil electrons, which becomes appreciable, does not change very much with the photon energy (σ_a in Fig. 1). The range of the secondary electrons is large in comparison with the grain size of the emulsion. Furthermore, the latent image formation will no longer be caused only by the photons absorbed in the photographic emulsion. With decreasing wave length, secondary radiation emitted from the film holder and film base becomes more and more important. The result is an intensifying action of near surroundings which has been used quite frequently in radiographic work with super-voltage X-rays (*15*, *16*) or γ-rays (*17*).

Intensifying Action of Lead

The nature of the intensifying action of lead screens has been investigated recently with the γ-rays of radium by Harrington *et al.* (*17*).

Secondary electron emission of a thin sheet of lead in front and behind the film was determined by totally removing the radiation originating in each screen. By also eliminating the effect of any other surrounding material (including air), the interaction of the γ-rays with the film alone could be measured. The electrons emitted from the front screen were prevented from reaching the emulsion by separating the lead sheet from the film and applying a magnetic field in the region between the screen and the film. This field prevents electrons ejected from the screen from reaching the photographic emulsion. Finally, the back screen was replaced by an evacuated space sufficiently long to assure a negligible effect from backward moving electrons and radiation from the walls of the container.

With Kodak non-screen film, a front lead screen 0.003 in. thick, and a back screen 0.02 in. thick, Harrington *et al.* obtained the following results with radium γ-rays:

Of the film blackening that occurs, 52% is caused by electrons ejected from the front screen. Nearly 48% of the film blackening results from electrons from the back screen. The blackening of the film due to the absorption of the γ-rays by the film itself is small, about 4%. The accuracy of this value is not very high, since it is about the limit of the accuracy of the experimental method. Furthermore the thickness of the front lead screen employed was much smaller than the range of most secondary electrons produced by the radium γ-rays in lead. An increase of the front screen thickness, however, would change the results only slightly, since before reaching the lead front screen the γ-rays first had to penetrate a thick piece of brass, which also emitted electrons.

From these results it follows that the direct action of high-energy photons on photographic material is negligible compared with the effect of the electrons emitted from lead foils. These are the only data available so far. Nothing is known about the interaction of X- or γ-rays of various energy with the film directly, in comparison with the action of the secondary electrons emitted from materials other than lead.

The comparison made by O'Connor (*16*) between an exposure of a naked film in complete darkness and one covered by various film holders does not give any additional information. A film surrounded by air only is exposed also to the secondary electrons originating in the air. Because of the low density of air the production rate of secondary electrons is small, but their ranges are large. Therefore the number of secondary electrons present per gram of material is the same in air as in any other low-Z material, if there is equilibrium between the electrons leaving and entering a certain volume of the material. In most cases, however, the width of the photon beam is small in comparison with the ranges of the electrons in air. Many electrons will escape from the path of the beam, but no electrons are scattered into this area, since no electrons are produced outside the beam. Therefore a dense material, shortening the electron ranges, shows a certain intensifying effect in comparison with exposures made in air (*16*).

Calibration of Film Response

The difference in photographic response of a film covered by lead or Masonite to X-rays of different maximum energy has been investigated in this laboratory. Masonite was chosen as the low-Z material because it has about the same density and composition

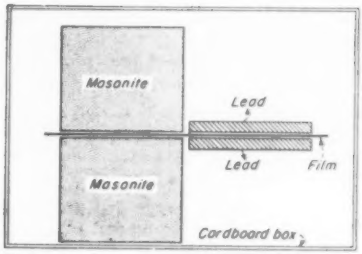

FIG. 3. Arrangement of photographic film strip covered partially by lead and Masonite and enclosed in a light-tight cardboard box for exposure to X-rays of various energies and intensities

as tissue and is therefore frequently employed as phantom material.

The X-rays were produced by electrons with energies of 0.5–3.0 Mev, accelerated by a Van de Graaff generator and absorbed in a gold target 3.2 mm thick. Including the water cooling, the X-rays had to penetrate an inherent filtration equivalent to 6.8 mm of lead. During the experiments additional lead filtration, corresponding to about two half-value layers, was always employed to reduce the relative intensity of the low-energy part of the X-ray spectrum. The secondary radiation from the lead was absorbed by additional filters of low-Z material.

For the measurements with a 10-Mev X-ray spectrum, the betatron of the U. S. Naval Ordnance Laboratory at White Oak, Md. was employed.* The upper limit of this X-ray spectrum was found to be at 11 Mev (*18*). From the published spectral distribution, the average photon energy is found to be about 4.1 Mev. But a total filtration of 2.5 cm of lead, corresponding to two half-value layers, was employed for these measurements. The actual average photon energy was somewhat higher than 4.1 Mev.

* The author is indebted to the U. S. Naval Ordnance Laboratory and to Mr. D. T. O'Connor for having the betatron made available for this investigation, and to Mr. K. A. Wright for having performed these measurements.

The experimental arrangement of the film exposed to the X-rays is shown in Fig. 3. Part of a strip of X-ray film was covered on both sides with lead; the rest of the strip was covered with Masonite. The thickness of the covering was greater than the range of the secondary electrons in that material. The surface of each material nearest to the X-ray target formed a plane with an edge of the film. The whole arrangement was placed inside a light-tight cardboard box so that the X-rays were incident parallel to the film surface, that is, perpendicular to the plane of Fig. 3.

The greatest blackening always appeared close to the film edge, where the primary and secondary radiation were in equilibrium. This depth changes with the photon energy, but it was always large enough for convenient measurements of the film density and small enough to neglect the X-ray absorption.

Kodak industrial X-ray film was employed and developed for five minutes with Kodak Liquid X-Ray Developer at standard temperature and slight agitation every minute. Since the

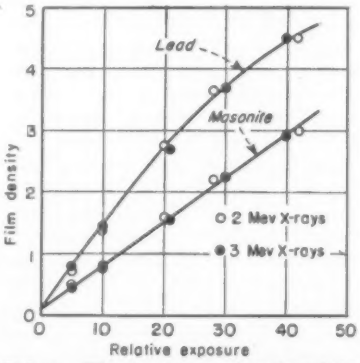

FIG. 4. Film density as a function of the relative exposure for films covered with Masonite and lead irradiated by 2- and 3-Mev constant-potential X-rays (Type A film, Kodak Standard Liquid X-ray Developer; film density includes base fog)

reciprocity law is valid for X-ray exposures (*14*), the product of beam current multiplied by exposure time is a measure of the relative dose delivered to the film. The readings of a Victoreen r-chamber next to the film box were used as controls.

Experimental Results

For the X-ray film covered with Masonite, the relation between density and relative exposure is linear (Fig. 4), in agreement with the results obtained by O'Connor (*16*). It was found that the density was independent of the X-ray energy (between 0.5 and 10 Mev) and only a function of the radiation dose. This could be expected, since all secondary electrons in low-Z materials are Compton electrons. The air dose is therefore proportional to the dose in any other low-Z material for this energy interval.

When aluminum was substituted for the Masonite, the results remained unchanged.

The density vs relative exposure obtained with 2- and 3-Mev X-rays for that part of the film strip covered by lead is also shown in Fig. 4. If the two curves of Fig. 4 are plotted on a logarithmic scale instead of a linear one, the more conventional H-and-D curves or characteristic film curves are obtained (Fig. 5). Since the difference in relative exposures for the two curves is constant for all densities, they allow a direct comparison of the different response of the photographic emulsion to the electrons emitted by the two materials.

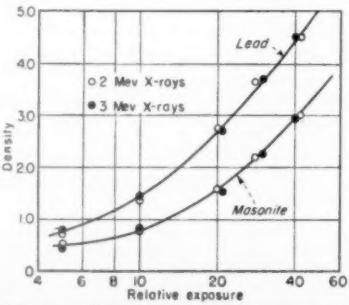

FIG. 5. Characteristic film curves for Kodak Type A film. Relative exposure plotted on a logarithmic scale for the data of Fig. 4

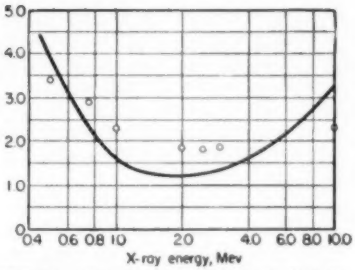

FIG. 6. Comparison of the relative intensifying action of lead and a low atomic number material surrounding Type A film irradiated by X-rays of seven different maximum photon energies between 0.5 and 10 Mev. Calculated curve taken from Fig. 2

The relative intensifying action of lead, compared with that of a low-Z material covering the film, has been determined in this way for seven different X-ray energies; the values obtained are given in Fig. 6. The experimental results are plotted for the maximum energies of the X-ray spectra, since the average photon energies are not well known. The intensifying action of lead has been found to be the same for X-ray energies between 2 and 3 Mev. The photoeffect increases the electron emission of lead at smaller photon energies in comparison to that of a low-Z material, while the pair production becomes noticeable at higher energies.

The curve in Fig. 6 is the same as that shown in Fig. 2, comparing the electron emission of lead and a low-Z material exposed to the same homogeneous radiation. In contrast to the relative counter efficiencies, no complete agree-

ment between the data obtained with the photographic material and the calculated ones could be expected for several reasons. The effective X-ray energy is actually much smaller than the maximum energy for which the experimental values have been plotted. The film density depends also on the energy of the secondary electrons emitted from the different materials, while the calculated curve compares only their relative numbers. Finally, it has been assumed so far that the interaction of the X-rays with the film directly is negligible. For the low photon energies, however, a certain fraction of the photographic density of the film covered with Masonite will not result from the electrons emitted by that material. Therefore the ratios with 0.5- and 0.75-Mev X-rays are rather low.

With monochromatic γ-rays instead of the continuous X-ray spectra, the photographic action of photons of various energies on emulsions covered by different materials could be obtained with better accuracy.

Conclusions

From the present investigation it follows that calibration of photographic material for high-energy (0.5–10 Mev) photon radiation is possible. The photographic emulsion has to be surrounded by a sufficient amount of material to secure equilibrium between the primary and secondary radiation. When the film is covered by a low atomic number material, a linear calibration of the film response can be obtained independent of the photon energy over a wide interval. Therefore, the radiation field of supervoltage X-rays in a phantom can be investigated by the photographic method, but this is not possible with low-energy radiation.

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A Simple Differential Pulse-Height Analyzer

This article describes an analyzer circuit suitable for use with scintillation counters. Multi-channel equipment can be easily built. Discriminator dead-time is about 1 μsec.

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THE ADVENT of the scintillation counter and its use as a proportional device (*1, 2*) has created a new demand for multi-channel and single-channel differential pulse-height analyzers. Several discriminators and differential discriminators have recently been described (*3, 4, 5*). These have been designed invariably for use with proportional counters. Because of the rapid action of scintillation counters, certain features are not essential for such use. Another analyzer depends on the principle of electron beam deflection (*6*).

In the proportional counter, because of the relatively long duration of the pulse, it is necessary to employ somewhat elaborate circuits to prevent the acceptance by the analyzer of a pulse which occurs before the previous pulse has decayed. This would not be necessary if the counting rates were low. But in order to obtain reliable results, high counting rates are desirable, not only to improve the statistics but also to avoid error caused by slow drift in the equipment. In the scintillation counter the pulse duration is of the order of one-thousandth of that in the proportional counter. What may be a high counting rate for the proportional counter may well be a low rate for the scintillation counter.

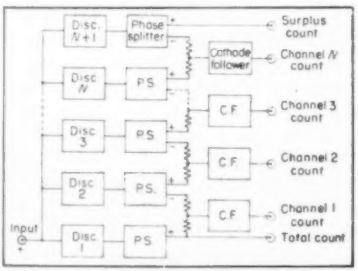

FIG. 1. Block diagram of analyzer

Circuit Description

The analyzer, shown in block diagram form in Fig. 1, is very simple; it consists of only three tubes for each amplitude level. The over-all arrangement is quite conventional. The discriminators employ a conventional type of two-tube trigger circuit. The anticoincidence circuit for each channel consists of a phase-inverter and two resistances. A cathode-follower is made available by using one-half of the phase-inverter double-triode. Thus, for a ten-channel differential discriminator, a total of 33 tubes is necessary, exclusive of power supply and scalers.

The discriminator circuit, shown in Fig. 2, consists of a modified Schmitt trigger circuit, in which there is no d-c coupling. Instead, capacitive coupling is used between the anode of the first tube and the grid of the second. The two tubes can then be biased off the same potentiometer network. This has the advantage that a separate high-stability resistance chain is not required in each discriminator level. The cou-

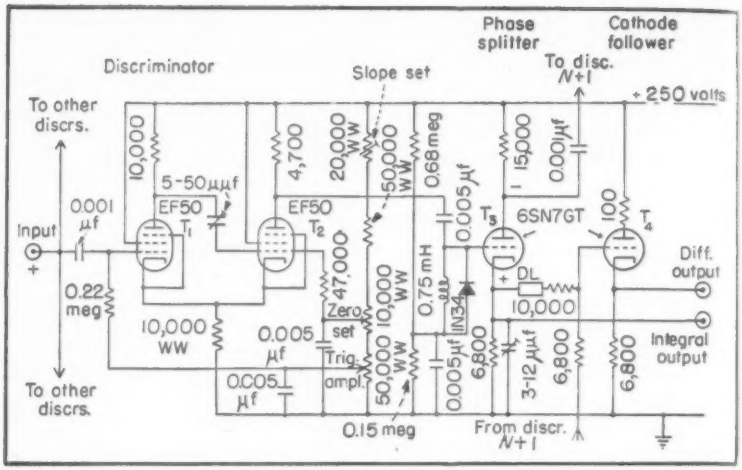

FIG. 2. Circuit diagram of a single level

pling capacitor should be made no larger than is necessary to give uniform output pulses.

Type EF50 tubes have been used, but 6AC7 or, better still, 6SH7, tubes may be used with little or no change.

The first tube T_1 of the pair is biased beyond cut-off. The minimum amplitude of positive-going pulse necessary to trigger the circuit is equal to the value of the bias voltage.

The resultant positive-going pulse appearing at the anode of the second tube T_2 is shaped by passing it into an inductance of about 0.75 mh shunted by a germanium diode, type 1N34. A positive pulse of about 0.5 μsec duration is obtained. This pulse is passed to a phase-splitting triode (half 6SN7GT) whose anode load is about twice its cathode load.

The positive-going pulse from the cathode of the phase splitter is fed into the 10,000-ohm end of a resistance chain consisting of two resistors—10,000 and 6,800 ohms. To the other end of this chain is fed the negative-going pulse (if any) from the anode of the phase-splitter of the next higher level. The junction of the resistors is connected to the grid of a cathode-follower, the spare half of the 6SN7GT tube in one level being used for this purpose.

If a pulse triggers two adjacent discriminators, the signal appearing at the junction of the 10,000- and 6,800-ohm resistors will be mainly negative-going. In practice, cancellation of the two pulses is not complete, and a small positive-going residue is obtained. This happens because the two discriminators do not trigger at exactly the same instant, the lower level firing slightly before the higher level, owing to the finite rise time of the incoming pulse. A small delay line (*DL* in Fig. 2) might be used to delay the cathode pulse, but it has been found possible to use the instrument without this. To reduce the effect, the anode load of the phase-splitter was purposely made larger than the cathode load. Also, the two resistances used in the anticoincidence chain were made to have different values.

If, on the other hand, only the lower

level is triggered, a positive-going pulse will be obtained. By adjusting the scaler discriminator level to a suitable value, all unwanted pulses can be excluded. The output voltage is approximately 6 volts and the residue for simultaneous pulses in two adjacent levels is about 2 volts, so that discrimination is not difficult.

By making a direct connection between the cathode of the phase-splitter and the grid of the cathode-follower through the 10,000-ohm resistor, a suitable grid potential is obtained.

For multi-channel operation the bias voltages for all the discriminators may be obtained from one potentiometer chain. The "zero-set" potentiometers may all be placed in parallel, and the 50,000-ohm "trigger-amplitude" potentiometer, supplying the bias for T_1, may have a number of fixed taps on it to supply the bias voltages for the various levels. The values of some of the components of the chain will have to be altered, but operation will be satisfactory, provided the positive end of the trigger-amplitude potentiometer can be set at + 100 volts relative to ground. The bias for T_2 (obtained from the zero-set control) should be variable between + 100 and + 120 volts. The remainder of the potential divider must drop 130 volts for a 250-volt supply.

Operation

To set up the equipment, the following procedure was adopted:

1. Adjust the 20,000-ohm resistance ("slope set") to give a suitable voltage drop across the trigger-amplitude control (100 volts is suggested).

2. Set the trigger-amplitude control at, say, + 90 volts and pass in pulses of about 10 to 15 volts. Adjust the variable coupling capacitor between T_1 and T_2 until uniform output pulses are obtained as the input pulses are varied above the threshold level. The capacitance should be as low as possible.

3. Set the trigger-amplitude control at + 100 volts. Adjust the zero-set control so that the trigger circuit is just below the threshold of free oscillation. For this adjustment there should be no connection to the input of the discriminator from auxiliary equipment, as noise may cause an error in the setting.

4. Adjust the capacitor in the cathode lead of the phase-splitter to make it aperiodic.

After adjustment, the amplitude of a triggering pulse will have a value equal to or greater than the voltage drop across the upper part of the trigger-amplitude potentiometer. Pulse distributions up to 100 volts may thus be analyzed.

In using this circuit, a highly stabilized d-c supply with a battery reference was used. The heater supply was taken from a stabilizing transformer. By this means, drifts were kept down to 0.1 volt or less in a day. A secondary battery could be used for heater supply where higher stability is required. Where channel widths of one volt or less are required, it is better to use a single-channel rather than a multi-channel arrangement, as drifts in adjacent channels in the latter might well occur in opposite directions. This could cause appreciable errors.

A high-impedance voltmeter may be used to read the channel width directly by connecting between the appropriate tapping points.

Negative output pulses may be obtained easily by putting an anode load in the cathode-follower stage and using it as a phase-inverter.

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The Birmingham Proton Synchrotron

Designed to accelerate protons to an energy of 1 Bev, this machine has been under construction at Birmingham University in England for over three years. It is thoroughly described here

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THE PROTON synchrotron concept grew out of the difficulties, both economic and technical, preventing the extension of cyclotron particle energies to higher values. With iron-cored magnet-type accelerators, increased energy requires increased radius. This makes a ring-type magnet very attractive for very high energies. The relativistic increase of mass with protons up to 1,000 Mev energy makes it impossible to accelerate them at constant angular velocity; frequency modulation of the accelerating r-f field, as in the synchrocyclotron, is necessary. By a further extension of the range (and accuracy) of the frequency modulation, the particles can be accelerated on a fixed orbit. This necessitates a rising magnetic field to keep the orbit radius fixed.

In the proton synchrotron both the frequency and the magnetic field are increased in such a fashion that the orbit radius is nearly constant. In order that the orbit should be confined within an annulus of small cross section, it is necessary for the radio frequency to be very accurately related to the rising magnetic field.

In several other respects the proton synchrotron presents technical difficulties which either do not exist or are not

Magnet with diameter of 32 feet is main component of the synchrotron. Design data for it and other components are tabulated below

General Properties

Maximum proton energy	1.3 Bev
Period of Acceleration	1 sec
Initial Energy per rev. (mean)	226 ev
Total number of revolutions	7×10^5
Radius of stable orbit	450 cm
Distance travelled	120,000 miles
Final energy spread	0.04%
Total possible number of particles	10^{12}
Average period of phase oscillation	250 μsec
Total number of phase oscillations	4,000
Available aperture for particles	35×10 cm

Magnet

Value of n	0.7
Maximum field strength	15,000 gauss
Average rate of rise of magnetic field	15,000 gauss/sec
Magnet gap (mean)	21 cm
Accuracy obtained in gap height	$\pm$0.005 in.
Radius of magnet	16 ft
Thickness of laminations	0.5 in.
Number of laminations	2,400
Total weight	810 tons
Thickness of pole tip laminations	0.125 in.
Number of pole tip laminations	16,000
Number of exciting turns	24
Peak current	12,000 amp
Peak voltage	1,100 volts
Rms current	4,600 amp
Energy stored (peak)	7,000 kw-sec
Magnet time constant	10 sec
Duty cycle	1 sec in 10
Dissipation in coils	182 kw (mean)
Dissipation in magnet	50 kw (mean)
Cooling air rate	15,000 ft^3/min

Motor Generator

Peak voltage	1,100 volts
Peak current	12,000 amp
Speed	500 rpm
Driving power	1,500 hp
Weight of flywheel	36 tons
Energy stored	55,000 kw-sec
Energy dissipation	500 kw

R-f System

Initial frequency	344 kc
Final frequency	9.6 Mc
Working voltage on Cee (rms)	240
Maximum available voltage for Cee (rms)	480
Angular length of Cee	96 degrees
Mean phase angle (240 volts)	22 degrees
Mean phase angle (480 volts)	11 degrees

Injection

Injection energy	500 kev
Magnetic field at injection	227.3 gauss
Effective time over which particles are accepted	100 μsec

nearly as serious in other types of accelerators.

The first known proposal for the construction of a synchrotron for accelerating electrons or protons to energies above 1 Bev was contained in a memorandum from M. L. Oliphant at Birmingham University to the Directorate of Atomic Energy in the Department of Scientific and Industrial Research in September, 1943 (*1*). The first published proposals were made by Veksler (*2*) in 1944, and McMillan (*3*) in 1945. These latter papers gave some theoretical treatment of the stability of the particle orbits. This theory has been examined in great detail in a large number of subsequent papers, of which the most important are by Bohm and Foldy (*4*), Frank (*5*, *6*), Saxon and Schwinger (*7*), Rabinovich (*8*), Dennison and Berlin (*9*), Gooden, Jensen and Symonds (*10*), and Twiss and Frank (*11*). Important aspects of the theory

will be summarized here before detailing the design and construction of the synchrotron.

Particle Oscillations

Betatron Oscillations. The success or failure of the proton synchrotron hinges on the behavior of particles that are disturbed from the ideal orbit and/or the ideal stable phase with respect to the accelerating r-f voltage. Particles possessing the correct energy for the ideal orbit and diverted from it by some means will describe free oscillations about that orbit, provided the magnetic field falls off with radius in accordance with the law $dH/H = -ndr/r$, where H is the field at radius r and dH is the change with change of radius dr. These oscillations are not connected with the radio frequency but are the well known betatron oscillations which occur in all magnetic resonance accelerators. Both vertical and horizontal free oscillations of this type can occur with frequencies $f\sqrt{n}$ and $f\sqrt{1-n}$ respectively, where f is the frequency of revolution of the particle in its orbit.

In any magnetic accelerator an initial oscillation, either vertical or horizontal, decreases in amplitude with increasing energy. The amplitude varies in proportion to $(Hr\sqrt{n})^{-\frac{1}{2}}$ for vertical and $(Hr\sqrt{(1-n)})^{-\frac{1}{2}}$ for horizontal oscillations. In the proton synchrotron both r and n remain substantially constant, and both oscillations diminish as $H^{-\frac{1}{2}}$. In practice, however, free oscillations are being produced at all times during the accelerating cycle because of scattering by the residual gas. At the lower energies oscillations build up from this cause more rapidly than $H^{\frac{1}{2}}$ unless the gas pressure is very low.

The loss of particles by increase of oscillation until collision with the vacuum chamber wall has been examined by Courant and Blachman (*12*). In the case of the Birmingham machine a pressure of 10^{-5} mm Hg would give rise to intolerable loss of particles; a pressure better than 10^{-6} mm Hg should be achieved to keep the loss from this cause unimportant.

Phase Oscillations. In addition to free oscillations, particles in a synchrotron undergo oscillations of phase relative to the accelerating radio frequency. Associated with these phase oscillations are additional radial oscillations which have the same period and are approximately 90 degrees out of phase.

This is strictly true only for oscillations of small amplitude. For large amplitudes the variation of phase and radius with time differs considerably from a sine function; however, zero radial displacement still corresponds to maximum or minimum phase displacement and vice versa. These oscillations are superimposed on the free oscillations and are independent of them.

The period of the phase oscillation in the Birmingham synchrotron varies from about 200 to 300 μsec during the period of acceleration. The number of complete oscillations is about 4,000. As the particle energy increases, the radial oscillation amplitude associated with a phase oscillation diminishes approximately as H^{-1}. In the case of the electron synchrotron, relativistic mass increase causes the phase-oscillation amplitude to diminish. In the proton synchrotron, certainly up to 1 Bev, the equivalent effect is negligible. For this reason artificial damping of the phase oscillations is desirable to ensure that oscillation build-up cannot occur through any cause. The limits of phase swing for which an oscillation is possible in the Birmingham machine are approximately 140 degrees in one direction and 90 degrees in the other, relative to the stable phase. Excursion outside these limits leads to instability, and particles doing so continue to change phase indefinitely and cease to be accelerated.

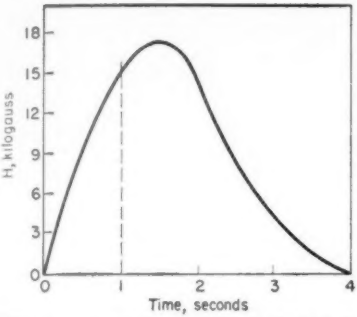

FIG. 1. Variation of magnetic field with time

The particles accepted at injection are, in general, distributed more or less uniformly over this range of phase angles. Errors in the radio frequency and rapid frequency or phase jumps can cause the oscillations to build up, leading to loss of some of the particles. To prevent this the ends of the accelerating electrode are cut at an angle so that particles experience different accelerating forces at different radii. This contributes a large damping term to the phase-oscillation equations and causes phase oscillations to be damped at a very rapid rate.

General Features

The choice of parameters in the design of a proton synchrotron involves a very difficult balance between many factors. Reduction in size, and hence cost, can only be achieved at the expense of increased technical difficulties in regard to magnet, r-f equipment and vacuum-system design. The compromise adopted has been weighted by the very considerable difficulties, both economic and practical, in launching a project of this magnitude in England immediately after the end of the war.

The principal data relating to the synchrotron and its component parts are summarized in the table on page 31. For design purposes a maximum particle

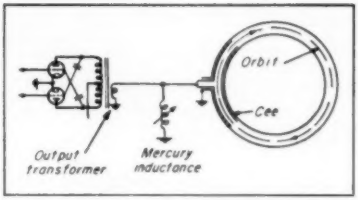

FIG. 2. Accelerating electrode (Cee), tuning inductance and output stage of power amplifier

energy of 1.3 Bev was chosen. The final value obtained will depend on magnet saturation and should be over 1 Bev.

Injection. The particles are injected at 500 kev into an orbit on the median plane just inside the outer limit of usable magnetic field. They are allowed to spiral inwards as the magnetic field rises, until the vacuum chamber is filled with particles. At this instant the radio frequency is switched on. All particles within the phase limits, whose radial excursions due to free and phase oscillation lie within the limits of the usable field, are captured and accelerated.

Two mechanisms for injection are provided and will be compared experimentally. In one the particles are injected at fixed energy; in the other, with energy rising linearly for a short period. Injection apparatus is detailed later in this article.

Every 10 seconds the magnetic field rises approximately linearly, as shown in Fig. 1, from the residual field to a value of 15,000 gauss in 1 second. The particles are injected when the field reaches a value of 227.3 gauss. For 500-kev particles, this corresponds to an orbit radius of 450 cm.

The particles receive increments of energy upon entering and leaving the accelerating electrode or "Cee," a drift tube occupying an arc of 96 degrees, as shown in Fig. 2. The average energy received per revolution

is 226 ev. The mean phase of the particle relative to the r-f voltage zero is 22 degrees. The r-f voltage on the Cee is 240 volts rms at all frequencies.

The relationship between frequency and magnetic field is

$$f = \frac{10.603H}{\sqrt{H^2 + 48.380}}$$

Here F is given in megacycles when H is expressed in kilogauss. This relationship is shown in Fig. 3.

Extraction. To extract particles at a given energy, the rising frequency can be locked when it reaches the required value. A constant frequency causes particles to spiral outwards.

For internal experiments this is all that is required. For external experiments the beam will be extracted by applying a sharp voltage pulse of some 200 kv or more to an extraction electrode. The pulse will be initiated by a trigger signal derived from proton nuclear magnetic resonance equipment set to operate at the predetermined magnetic field corresponding to a particle orbit adjacent to the extraction electrodes. (Nuclear resonance equipment is being used for all the magnetic field measurements.)

Construction of Magnet

The magnet consists of 800 tons of C-shaped low-carbon (0.1 %) steel plates assembled in the form of a circle with the gap outwards, as shown in the illustration at the beginning of this article. The shape of a plate is shown in Fig. 4. The thickness of the plates is ½ in. The plates are flat and touch at the inside edge. Above and below the gap on the outside half of the magnet, steel wedge plates are inserted after every pair of flat plates, as shown in Fig. 5.

The wedge-shaped spaces on the inner half of the magnet are used for forced air cooling of the windings. Air enters at the top of the magnet via these wedges, is diverted by baffles into

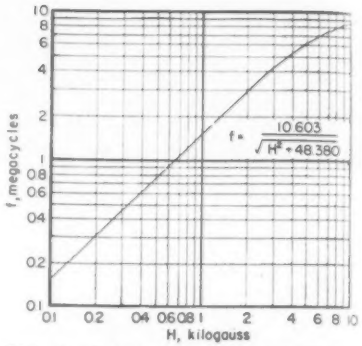

FIG. 3. Relation between accelerating radio frequency and magnetic field

the winding space, and then is drawn off at the bottom via the wedge spaces. A large axial-flow fan located in the hollow center of the magnet supplies 15,000 ft³/min of air.

The magnet plates are held together by bands of welds around the magnet on the inside, outside, and top edges of the plates. There are no welds within the gap or winding space, as these would short circuit the changing flux in the main gap.

The magnet pole tips are made of soft iron plates ⅛ in. thick, insulated by paint on each plate and paper between groups of plates. The average gap is 21 cm, and the value of n is 0.7. The pole-tip surfaces have been assembled with an accuracy in the gap dimensions

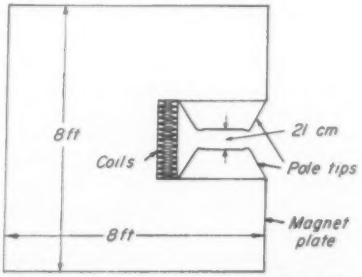

FIG. 4. Cross section of magnet plate

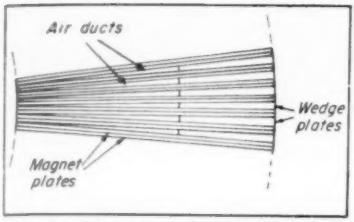

FIG. 5. Plan view of a section of the magnet showing pairs of flat ½ in. magnet plates angle spaced by short wedge plates

of better than ±0.005 in. To achieve this accuracy over the magnet diameter of 30 feet, a special water level gage was constructed. A telescope was used to observe the position of contact of a sharp point with a water surface. Two such gages connected by a horizontal water pipe were used to establish reference levels all around the magnet to within 0.001 in.

The magnet winding consists of 144 turns of stranded conductor (380 /0.036 in.) arranged in 6 parallel coils of 24 turns each. The magnet has an inductance of 0.1 henry and a resistance of 0.01 ohms. Its time constant is thus 10 sec. The maximum current through the winding is 12,000 amp, for which the stored energy is 7,000 kw-sec. The mean dissipation in the magnet iron is 50 kw; in the coils, 182 kw.

The magnet was built on a reinforced concrete raft resting on 30 piles driven down to firm rock. The magnet plates rest on bridge rails laid on the concrete surface. Thermal expansion and contraction of the magnet will cause the plates to slide across these rails. A reinforced concrete anchor ring around the base of the magnet prevents any excessive sideways movement.

Generators

The magnet is excited by a pair of large d-c generators connected in parallel. The generators are driven at 500 rpm by a 1,500-hp 11-kv three-phase

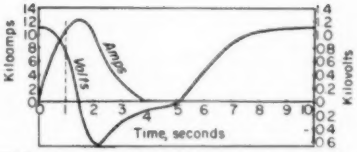

FIG. 6. Variation of generator voltage and magnet current with time over a complete synchrotron cycle

50-cycle induction motor. The load peak every 10 sec is carried largely by a 36-ton flywheel coupled to the generators.

The total energy stored in the rotating machinery is 55,000 kw-sec. Average power dissipation is over 500 kw, most of which is due to windage.

Magnet Cycle

The generator voltage and current cycles are shown in Fig. 6. At the start of the cycle the voltage is stationary at 1,100 volts. The cycle is initiated when the variable r-f generator supplies a trigger pulse to a thyratron which strikes a type BK22 glass-bulb ignitron connected between the generator and the magnet. The striking of the ignitron is reproducible to 1 μsec.

The main switch pull-in coil is energized by another thyratron, also triggered from the r-f generator. The main switch consists of nine 600-amp contactors (continuous rating) in three banks of three, all connected in parallel. The operating time is 120 msec, and therefore the switch by itself could not be relied upon for anything like 1 μsec timing stability.

The ignitron is capable of passing currents rising to 1,000 amp, but the main switch is connected in parallel with it and is timed to close when the magnet current has reached 300 amp. Thus the ignitron arc is quenched when this current is reached and its average dissipation is quite trivial.

The main switch pull-in coil takes 70 msec to build up sufficient current

to start moving. Time of flight from this point is 50 msec. This makes it possible to protect the ignitron against the certain damage that would ensue if it were to strike and the main switch did not close. This protection is afforded by an additional contact on the main switch in series with the ignitor of the ignitron; this contact is not closed until the main switch is in motion.

The magnet current rises more or less linearly for 1 sec. During this time the particles are accelerated. The current continues up to 12,000 amp before the rise can be checked. Thereafter it is reduced and reaches zero about 4 sec after the beginning of the cycle. When the current approaches zero, the main switch falls out and isolates the magnet until the beginning of the next cycle.

The removal of energy from the magnet is effected by reversing the main generator voltage until the magnet current reaches zero. To reverse the generator voltage, it is necessary to reverse its field current. This current is provided by a field generator, the field of which is, in turn, provided by a pilot field generator. It is necessary to reverse the field of the pilot field generator about 0.25 sec after the beginning of the cycle to keep the magnet current from rising to too high a value. The machine time constants are so long that the main generator voltage is still nearly 800 volts by the end of the 1-sec cycle.

Five seconds after the beginning of the cycle, the generator voltage begins to rise to its maximum value of 1,100 volts. It has five seconds in which to reach this value and settle down before the start of the next cycle.

R-f Range and Accuracy

The useful half width of the vacuum chamber is about 3.3% of the mean orbit radius. At the highest energies, when the particle beam has contracted to a small radial dimension and the velocity approaches that of light, the permissible frequency error can approach this value. At injection the complete space is filled with particles, and any frequency error will cause some loss. On this basis the frequency tolerance for 10% loss of particles is $\pm 0.23\%$ and for 50% loss is about $\pm 1.1\%$. As the particle bunch contracts laterally because of increased energy and phase-oscillation damping, the permissible tolerance rapidly increases. Within a few milliseconds, the tolerance for 10% loss is up to $\pm 1\%$; thereafter it rises progressively to $\pm 3\%$ at 1 sec. The r-f system has been designed to achieve a frequency accuracy of $\pm 0.25\%$ at injection and immediately afterwards, and an accuracy throughout the remainder of the cycle sufficient to ensure no further loss of particles from this cause.

At injection the required frequency is 340 kc. At the highest energy it is 9.6 Mc. An error of 0.25% at injection is 860 cycles, which represents 1 part in 11,000 of the final frequency. It is clear from this that the system for producing the required frequency sweep must be capable of very fine adjustment, at least at injection.

When the design of the r-f system for the Birmingham synchrotron was started, it was planned to inject particles with an energy of 250 kev. In this case the required frequency range is larger still (39 to 1). When it was subsequently decided to inject at a higher energy, the wider frequency range was retained, as it would allow the machine to be used with either protons, deuterons or alpha particles by a suitable change in the frequency-magnetic field relation. With this in mind a frequency generator, wide-band power amplifier and accelerating-electrode matching and tuning system have all been designed to make this wide frequency range available with the requisite accuracy.

The rising magnetic field is defined

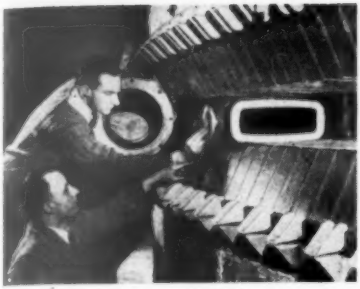

FIG. 7. Vacuum box section in place between pole tips. Pumping ports and a section of the vacuum manifold can be seen

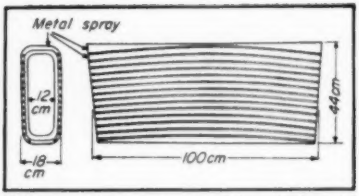

FIG. 8. Vacuum box section, 100 × 44 × 18 cm, showing the arrangement of sprayed copper conducting strips as used on the Cee sections

primarily by the generator voltage, the magnet air gap, and the number of turns on the winding. The only unreproducible factor is the generator voltage. The time constants of the main generator field control system are too long to permit accurate stabilization of the generator voltage in the time available. It is necessary, therefore, to take the magnetic field as it comes and make corrections via the radio frequency. This means that the frequency sweep generator cannot produce a fixed frequency-time relation, but must be controlled in some degree by the magnet field or current or by the generator voltage. The frequency control system was designed on the assumption that the generator voltage would be reproducible from cycle to cycle to within $\pm 2\frac{1}{2}\%$ and that the rate of change of field near injection is accurately proportional to this voltage.

Vacuum Box and Accelerating Electrode

The final details of the vacuum box and Cee system have not been settled at the time of writing. The following description covers the essential design features and final form of construction as envisaged.

The Cee shown in Fig. 2 must be fitted in the magnet gap in such a way as to enclose as much as possible of the usable region without involving excessive r-f power losses. The voltage on the Cee gives rise to a substantial r-f current along the outside of the Cee, together with an equal return current on the adjacent magnet pole faces. The Cee and the pole faces form a transmission line whose characteristic impedance is a few ohms. To reduce r-f losses, the pole faces will be covered with copper 0.002-in. thick, sprayed on in strips 1-in. wide and separated $\frac{1}{16}$ in. For insulation purposes and to cover the gaps between laminations and give a good surface for spraying, the pole surfaces will be completely covered with adhesive tape (2 in. strips).

The copper is in 1 in. strips to minimize eddy-current interference with the rising magnetic field. A sprayed film is not as good as metal foil as regards conductivity, but a thickness of 0.002 in. gives satisfactory r-f performance.

The Cee will be formed in a similar fashion by spraying copper strips on the outside of some of the vacuum box sections. The vacuum box consists of a number of sections with a 3 cm wall thickness similar to the stone box between the magnet pole faces in Fig. 7. A complete set of 30 stone boxes has been constructed with satisfactory mechanical properties but with an unduly high leak rate caused by porosity. These can be rendered satisfactory by beeswax impregnation, but a much more attractive solution using electrical

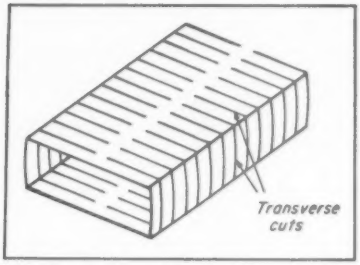

FIG. 9. Section of Inconel liner inserted in vacuum box to remove stray surface charges. Transverse cuts reduce eddy current disturbance of magnetic field

porcelain has been developed recently. It is probable that the final box will consist of 60 of the porcelain sections.

Electrical connections between vacuum box sections will be obtained by carrying the metal spray over the edges of the ends, as shown in Fig. 8, and fitting thin gaskets of insulating material with spring contacts at each junction. These contact gaskets will be outside the rubber vacuum-seal gaskets.

A metal lining is required inside the vacuum box to remove stray charges which would otherwise accumulate on the inside wall. Quite small charges collecting on very small exposed surfaces have been known to produce strong electrostatic fields and ruin the operation of other accelerators. A fabricated metal liner, constructed from thin sheets (0.018 in.) of a high resistivity nonmagnetic alloy (Inconel) and cut as shown in Fig. 9 to reduce eddy-current disturbance of the magnetic field, has been considered more satisfactory than the metallizing techniques.

At the ends of the Cee the liner is broken and a connection made through the vacuum-box wall to the ends of the copper strips. These strips together with this section of the liner form the complete Cee. The rest of the liner beyond the ends of the Cee will be connected through the box walls to the ends of the Cee's earthed screen on the pole faces. All connections through the wall are made by flat sheets of metal with rubber-ring vacuum seals on each side. In each case, inside the box, direct connections will be made to the liners, but, outside, small resistors will be connected to each of the longitudinal strips. If direct connections were made to the strips they would close the paths for eddy-current flow and annul the advantage of using strips instead of sheet.

The region between the liner and the Cee conductors is free from r-f fields; consequently the r-f properties of the stone or porcelain are unimportant. Some dielectric loss occurs at each end of the Cee, but even with a poor power factor it is not serious.

The minimum clearance between the outside walls of the vacuum box and the pole faces is 1.5 cm. The transmission line formed by the Cee and the pole faces has a characteristic impedance of 7.6 ohms and a propagation velocity equal to the velocity of light. At 9.6 Mc the 96 degree Cee is about 90 electrical degrees in length. If the

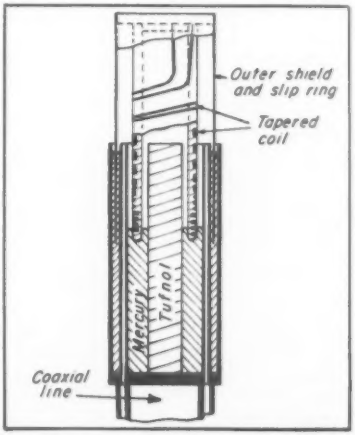

FIG. 10. Variable inductance for tuning the accelerating electrode. A tapered coil is plunged into mercury and the inductance varied by a ratio of 1,000 to 1

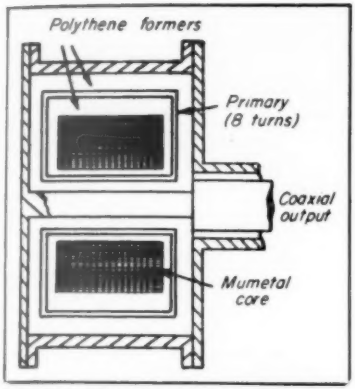

FIG. 11. Wide-band transformer matching the tuned Cee to the power amplifier. The case forms a single-turn secondary

Cee occupied 180 angular degrees, it would be very nearly a resonant half-wave line at 9.6 Mc and would be very difficult to match over the required frequency band. For this reason the Cee has been shortened. The r-f voltage is increased to give the same particle energy per revolution with the shorter Cee, but energy requirements remain about the same.

Mercury Tuning Inductance

The Cee impedance at its center varies from $-3.8j$ ohms at 9.6 Mc to $-194j$ ohms at 0.25 Mc, a ratio of 52 to 1. A tuning inductance to tune the Cee alone at all frequencies thus requires an inductance range of 40×52, or 2,080 to 1. Accurate tuning at low frequencies can be dispensed with and an inductance ratio of 1,000 to 1 is adequate. This is achieved with adequate accuracy by means of a tapered coil which is varied by plunging into mercury. See Fig. 10.

A tapered coil is wound on a Tufnol tube and dips into mercury in the inner pot. A cylinder dips into another cylindrical pot surrounding the first and acts as a slip ring completing the r-f current return path. The system is thus completely screened, and stray lead inductance is kept to a minimum.

The connection to the Cee is made via a very short coaxial line from the bottom of the inductance. The coil consists of several pancakes of five turns each at the bottom followed by a tapered spiral winding with increasing pitch and wire size, ending in a wide strip of metal at the top. Nickel is used for the winding and chromium plated stainless steel for the pots. The tuning accuracy is such that the impedance level is raised from a minimum of $-1.5j$ ohms (total of the Cee, mercury inductance, coaxial feeder, output amplifier and output transformer capacitances) to a minimum of $\pm 25j$ ohms.

Power Amplifier

The voltage required on the Cee for optimum results is 240 volts rms at all frequencies. The power amplifier was designed to be able to develop twice this voltage at the Cee ends at all frequencies. The reactive power in the output circuit at 9.6 Mc is thus about 75 kva. The peak power dissipated in the circuit is about 3 kw, most of which goes into core losses in the matching transformer.

The output amplifier consists of a pair of 1-kw anode dissipation triodes, type 3J/160E, operated with 3 kv on the anode and zero grid bias. The 1-sec-in-10 duty cycle keeps the anode dissipation small. The output stage is preceded by a broad-band amplifier culminating in a driving stage using two 3J/160E tubes with 3 kv, operated in class A.

The output transformer, Fig. 11, has an eight-turn center-tapped primary wound on a mumetal toroidal core. The secondary has a single turn and is formed by the case. The primary winding and core are supported by polythene formers, and the winding is

made with a void-free polythene-covered wire coated over the polythene with a thin layer (0.0005 in.) of polyvinylchloride. The transformer is vacuum filled with paraffin oil; the polyvinylchloride prevents the oil from affecting the wire insulation. The mumetal core is made up of 14 pancakes each ¼ in. thick consisting of mumetal tape 0.0008 in. thick.

R-f Generator

The required frequency sweep is generated by beating a fixed oscillator of 35 Mc with an oscillator varying from 34.66 to 25.4 Mc. The required accuracy at injection is equivalent to a change in the frequency of one of these oscillators of 1 part in 40,000. This calls for a high order of stability in the fundamental oscillators—at least ten times better than this figure.

Because of the wide frequency ratio (39 to 1) and the high upper frequency (10 Mc), coupled with the considerable accuracy (0.25%) required at the lowest frequency, it was not considered feasible to develop a more direct means of generating the required frequency law by using the magnetic field or the magnet current to control the characteristics of an electronic device or the r-f properties of a dielectric or ferromagnetic medium.

The fixed and variable oscillators both use coaxial quarter-wave lines as the basic tuning elements. The outer conductors of the lines have a diameter of 8 in., and the lines are each 7 ft in length. The variable oscillator is tuned by a capacitor at the open end.

The capacitor is shown in Fig. 12. It consists of a single large aluminium vane (24 in. diameter) meshing with a small stator attached to the end of the coaxial inner conductor. The vane is cut in a series of steps so that a step enters the stator each time the preceding step leaves.

This construction was adopted because of the very high mechanical stability required. The period of rotation is 1.2 sec. With the construction shown, an accuracy of 0.25% at injection corresponds to a meshing accuracy of 0.0014 in. and a timing accuracy of 32.5 μsec. Tolerances at least 10 times tighter than these figures have been adopted where possible. Bearing slack is no more than 0.0001 in., and variations caused by this slack are minimized still further by the initial shape of the capacitor vane. This shape also minimizes effects caused by differential thermal expansion of the rather large members. Expansion effects are reduced still further by the use of materials with similar expansion coefficients, by temperature stabilization to ±0.5° C, and by introducing similar geometry in the fixed and variable lines as far as possible.

The capacitor vane is completely enclosed in a machined casting. The clearance between the vane and the casting is 0.5 mm at top and bottom.

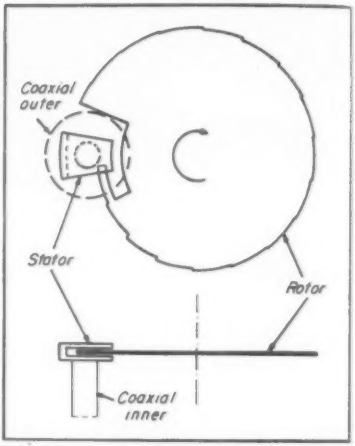

FIG. 12. Variable capacitor for generating the accelerating radio frequency. The capacitor is attached to the open end of a high-Q coaxial line. The stepped rotor is coupled capacitively to the coaxial outer

This provides the by-pass capacitance to the coaxial outer conductor. The rotor bearing is isolated at radio frequencies by surrounding the spindle just outside the bearing with a toroidal iron-dust core ($\mu = 50$, $Q = 6.7$ at 35 Mc). This acts as a single-turn choke and gives a very adequate degree of isolation at the expense of a small deterioration in system Q.

The circuit of each oscillator consists of a loop coupling out of the line into a cathode follower and thence to a pentode amplifier feeding into a reversed loop coupling back into the line. Miniature high-transconductance (g_m) 7-pin button-based tubes (Mullard type EF91) are used for the cathode follower and for amplifier applications.

The circuit is designed to achieve the maximum stability. Tests with the two oscillators beating together show that a relative stability of one part in a million will be approached over a reasonable period of time.

In the usual line-stabilized oscillator another tuned element is incorporated to prevent operation at the higher harmonics of the line In this case the addition of another variable tuned element was avoided by placing the two loops together at a point one-third of the way up the line from the short circuit This makes operation at the third (and probably higher) harmonic impossible. No trouble was obtained with this arrangement, whereas the oscillator alternated between fundamental and third-harmonic operation when the loops were placed at the bottom.

Capacitor Speed Control

The capacitor vane rotates at a speed which is accurately proportional to the synchrotron magnet generator voltage. The vane is driven through a 12-to-1 reduction gear servomotor which is capable of being switched rapidly. The motor is excited by a high-frequency (~1,000 cycle) voltage and establishes a desired torque within a millisecond of application of the control signal.

For speed control purposes the motor is switched to accelerate, retard or maintain speed for a period of 1 msec at a time. The sign and degree of control are determined every 10 msec, in effect, by a comparison of the speed of the vane with a suitably corrected reference speed, the correction being derived from the synchrotron magnet generator voltage.

To determine the capacitor speed accurately, 120 narrow strip mirrors are used, one placed every 3 degrees around the vane. As the vane rotates, the mirrors reflect light back into an optical system and produce sharp voltage pulses at the output of a photomultiplier.

The mirrors are set to an accuracy of 2 seconds of arc, and, in consequence, the voltage pulses are accurately timed to 2 μsec. The interval between consecutive pulses is 10,000 μsec if the vane is rotating at the speed corresponding to an accurate generator voltage.

Speed measurement is effected by comparing the time interval between pulses with a reference interval. When the generator voltage is correct this reference interval is derived from a 1-Mc pulser followed by a four-decade binary scaler. Each mirror pulse opens a gate allowing the scaler to work. After 10,000 μsec the scaler shuts the gate. The difference in time of the scaler output and the next mirror pulse is an accurate measure of the average speed of the disk over the 10,000-μsec period and is used to control the servomotor.

When the generator voltage is low, the gate between the 1-Mc pulser and the scaler is closed for a short period, delaying the output pulse from the scaler. This causes the servomotor to slow the vane. The period for which the gate is shut is decided by circuits in which the desired generator voltage, 1,100 volts, is subtracted from the

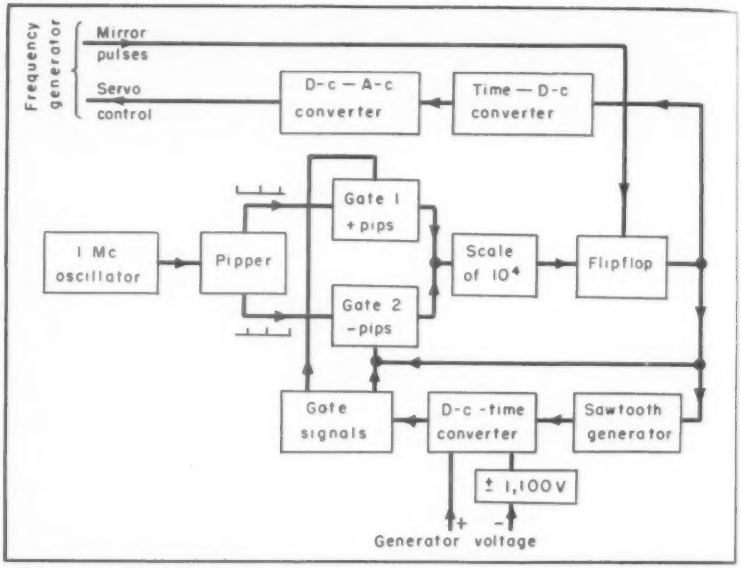

FIG. 13. Block diagram showing speed measurement, correction, and control units used to relate the variable capacitor speed to the magnet generator voltage

actual generator voltage. The difference controls a voltage-time converter giving the desired gate control waveform output.

When the generator voltage is high, the scaler must operate earlier. This is done by opening a similar gate which is normally closed and which allows another chain of 1-μsec pulses through to the scaler. These pulses are spaced midway between those in the first channel.

A block diagram of the speed control system is given in Fig. 13.

Particle Injection

Ion Source. An ion source of the Thoneman type (*13*), operating at 18 Mc and delivering a 1 ma current of over 80% protons, is used because of its inherently narrow energy spread. The r-f supply is pulsed on for about 0.3 msec and is fed to the source from an oscillator near ground potential. The power is fed along a parallel-wire transmission line consisting of series capacitors and shunt inductors, all enclosed in an oil filled tube which has a diameter of 7 in. A string of high-value resistors is used to divide the 500 kv evenly between the series capacitors. The other power required in association with the ion source is obtained from a belt-driven generator.

Injection System. The injection high voltage is derived from a 500-kv Philips set using cascade rectifiers. It is built up from a platform in a large pit (a cyclotron pit) next to the synchrotron in such a way that the high-voltage electrode is near the median plane of the synchrotron. The ion source is exactly on the median plane, and the paths of particles being injected lie on this plane.

The ion beam is accelerated through

three gaps formed by mild steel electrodes which shield it from the stray magnetic field. The beam is then allowed to "coast" along a tapering mumetal tube until it reaches to within a few centimeters of the magnet pole tips. Up to this point the beam travels in a straight line.

Emerging from the mumetal tube, the beam is guided between two circular arc electrodes until it reaches the injection orbit. A rising voltage between the two electrodes during the injection period keeps the beam on its correct path despite the rising magnetic field.

The horizontal width of the proton beam and the cone angle must both be kept very small if an appreciable fraction of particles is to be accepted into suitable orbits. The beam spirals into the center at about 3 mm per revolution when the accelerating radio frequency is not applied. Thus, if most of the beam is to miss the inside edge of the circular arc electrodes the next time it approaches the injection point, it is desirable that the beam width be small compared with 3 mm.

Particles entering at an angle to the correct orbit execute free betatron type oscillations which add to the proportion lost by hitting the injector. To keep loss due to these causes small, a total cone angle of 0.05 degrees would be desirable, together with a beam width of 1 mm. These figures are very difficult to achieve and are further aggravated by other errors such as inaccurate positioning or directing of the cone center line, and variations in the magnetic field. It is not yet possible to say what accuracy will be achieved.

Vacuum Pumps

The vacuum chamber is pumped by means of five 16-in. diameter diffusion pumps distributed around the synchrotron and feeding into a common backing line. Each pump has a nonmagnetic stainless-steel manifold which connects with three vacuum box sections. Each section has four outlet ports. The pumps have water-cooled baffles and liquid-air traps. The accelerator tube between the ion source and the injection point is differentially pumped to dispose of gas from the ion source.

Beam Monitoring

Equipment has been developed to portray on long persistence cathode-ray tubes the size and position of the proton beam during the acceleration period. It is hoped that this will facilitate testing of the machine.

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Chemical Syntheses of Radioisotope-Labeled Compounds—II

This is the second half of an up-to-date status report on the rapidly increasing activity in the field of chemical synthesis of compounds labeled with C^{14}, I^{131}, S^{35}, and P^{32}. Included is a list of compounds whose syntheses are being investigated under contracts from the Isotopes Division

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IT WAS NOTED in Part I of this status report* that its preparation was undertaken in view of the growing interest in chemically synthesized compounds labeled with radioisotopes.

General comments were offered on the availability of commercially synthesized labeled compounds and on the progress being made in establishing uniform nomenclature for all radioisotopic compounds.

The major portion of this report is essentially a compendium of almost all the radiochemical syntheses completed since the book "Isotopic Carbon" was published.† Although details with regard to particular techniques used are not included, the listing of the individual syntheses in the manner in which they are recorded here serves to indicate the general routes and procedures involved.

The report is comprised of twelve sections and is broken down into categories of the compounds of carbon-14 (sections *1–9*), iodine-131 (section *10*), sulfur-35 (section *11*), and phosphorus-32 (section *12*). Part I of the report contained sections *1–3* on the following carbon-14 compounds: *1.* Low Carbon Intermediates (C_1 to C_4); *2.* Acyclic Acids and Derivatives; and *3.* Amino Acids. Sections *4–12* follow below.

Preceding each section are a few highlights in the development of that particular group of compounds.

Following section *12*, on page 61, there are listed the compounds whose syntheses are being investigated under current contracts with the Isotopes Division.

* NUCLEONICS **7**, No. 3, 49 (1950). This report is a companion piece to the report on biochemically synthesized compounds [NUCLEONICS **7**, No. 2, 26 (1950)].

† "Isotopic Carbon," by M. Calvin, C. Heidelberger, J. Reid, B. Tolbert, P. Yankwich (John Wiley & Sons, Inc., New York, 1949).

4. CYCLIC COMPOUNDS

The Friedel-Crafts condensation using labeled acyl halides has resulted in a considerable number of labeled aromatic derivatives. Considerable effort has been expended and is currently being directed toward methods for the synthesis of ring-labeled benzene and its derivatives. So far, this synthesis has been successfully accomplished by the alkaline trimerization of pyruvic acid-

2-C^{14}, the condensation of ethyl acetate-1-C^{14} with the Grignard of pentamethylene dibromide, and a Diels-Alder condensation between maleic anhydride-2, 3-C_2^{14} and butadiene. Progress in this direction should soon provide a number of different routes to this important class of radioisotopic compounds.

The synthesis of labeled styrene has led to convenient commercial counting reference sources for carbon-14 work. The general availability of thin squares of labeled polystyrene plastic has just been announced by the AEC Isotopes Division. A 1¼-inch square of polystyrene sheet, 10 to 12 mils thick, contains approximately 0.6 microcurie of C^{14} and is priced at $5. The activity in terms of an average end-window G-M assembly is roughly 7,000 counts per minute (10 mm from a 1.5 gm/cm² end window).

NOTE: Asterisk (*) before the compound denotes reference appeared in "Isotopic Carbon," and a page number in the book is given for references unavailable elsewhere.

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*Acetophenone-carbonyl C^{14}. W. G. Dauben, J. C. Reid, P. E. Yankwich, unpublished work (page 200); also O. Neville, W. Brown, MDDC-1168

Aceto-1-C^{14}-phenone; 1-phenylglyoxal-1-C^{14}; mandelic acid (2-phenylglycolic acid-2-C^{14}). O. K. Neville, *J. Am. Chem. Soc.* **70,** 3499 (1948)

From acetic acid-1-C^{14} and benzene, a Friedel-Crafts condensation yielded aceto-1-C^{14}-phenone in 78% yield.

Selenium dioxide oxidation then gave 1-phenylglyoxal-1-C^{14}. The alkaline rearrangement of phenylglyoxal afforded mandelic acid, with no rearrangement of the carbon chain, in 62% yield.

Aceto-1-C^{14}-phenone. J. D. Roberts; Massachusetts Inst. of Technology.

Acetyl chloride-1-C^{14} was reacted with diphenylcadmium and acidified with H_2SO_4 to give a 24% yield of the desired product.

*p-Anisaldehyde (carbonyl-C^{14}). J. Reid, H. Jones, *J. Biol. Chem.* **174,** 427 (1948)

*p-Anisic acid (carboxyl-C^{14}). J. Reid, H. Jones, *J. Biol. Chem.* **174,** 427 (1948)

Anthranilic acid-7-C^{14} (carboxyl-C^{14}-o-aminobenzene). A. Murray, A. R. Ronzio, Los Alamos Sci. Lab. AECU-85

This synthesis involved the carbonation of the lithium Grignard of o-bromoacetanilide. Fresh butyllithium in ether at −30° C gave the desired lithium compound with o-bromoacetanilide. After the carbonation, the mixture was hydrolyzed and ether extracted. The yield was 50%.

Benzalaceto-1-C^{14}-phenone. J. D. Roberts, Massachusetts Inst. of Technology

Aceto-1-C^{14}-phenone was condensed with benzaldehyde in sodium ethoxide to give a 100% yield of product. The dibromide derivative was prepared in 77% yield by the addition of bromine.

Benzaldehyde-7-C^{14} (formyl-C^{14}-benzene). O. K. Neville, Oak Ridge Natl. Lab.

The carbonation of phenylmagnesium bromide was followed by lithium aluminum hydride reduction to phenyl carbinol-C^{14}. This latter compound was then oxidized to formyl-C^{14} benzene with chromic acid.

*3,4-Dimethoxybenzaldehyde (carbonyl-C^{14}). J. C. Reid, unpublished work (page 199)

Benzanthracene-5,6-C^{14}. C. J. Collins, J. G. Burr, Oak Ridge Natl. Lab.

This compound was synthesized utilizing the procedure of Collins, *J. Am. Chem. Soc.* **70,** 2418 (1948). Degradation studies revealed the positions of labeling.

Benzene-1-C^{14}; benzoic acid-1-C^{14}; toluene-1-C^{14}. M. Fields, M. Leaffer, J. Rohan, *Science* **109,** 35 (1949)

Ethyl acetate-1-C^{14} was synthesized in 85% yield from barium carbonate and reacted with the magnesium Grignard of pentamethylene dibromide.

A 60–70% yield of 1-methyl-1-hydroxycyclohexane-1-C^{14} was obtained. This was

then dehydrated with iodine in 80–85% yield to the methyl-substituted cyclohexene. Vapor phase dehydrogenation over platinized asbestos afforded a 90–95% yield of toluene-1-C^{14}.

Permanganate oxidation gave benzoic acid-1-C^{14} in 90–95% yield. Decarboxylation to benzene was accomplished in 85% yield using cupric oxide and quinoline.

Ethyl-2-C^{14}-benzene. G. Ropp, O. K. Neville, Oak Ridge Natl. Lab.

Aceto-2-C^{14}-phenone was catalytically reduced, using palladium on charcoal catalyst in the presence of perchloric acid, to ethyl-2-C^{14}-benzene in 50–60% yield.

***Benzoic acid.** W. G. Dauben, J. C. Reid, P. E. Yankwich, *Anal. Chem.* **19,** 828 (1947)

***p-Aminobenzoic acid.** A. Murray, W. W. Foreman, W. H. Langham, *J. Am. Chem. Soc.* **70,** 1037 (1948)

***3,4-Dimethoxybenzoic acid [-carboxyl C^{14}].** J. C. Reid, unpublished work (page 183)

Benzoic acid-1,2-C_2^{14}. R. F. Nystrom, Y. H. Loo, K. M. Mann, J. R. Allen, Univ. of Illinois

Phthalic acid-1,2-C_2^{14} was synthesized by a Diels-Alder condensation between maleic anhydride-2,3-C_2^{14} and butadiene.

The mercuric salt of phthalic acid was isolated and refluxed in hydrochloric acid at 140° C for three hours. Benzoic acid-1,2-C_2^{14} was subsequently isolated in 90% yield.

Benzoic acid-2,3-C_1^{14}. C. J. Collins, Oak Ridge Natl. Lab.

Phthalic acid, resulting from the oxidation of 2-methyl-1,4-naphthoquinone-8-C^{14}, was decarboxylated by heating at 325° C for 12 hours.

A quantitative yield of benzoic acid-2,3-C_1^{14} was obtained.

p-Chlorobenzil [1-(p-chlorophenyl)-2-phenylglyoxal-2-C^{14}]. O. K. Neville, Oak Ridge Natl. Lab.

Condensation of *p*-chlorobenzylmagnesium chloride with carbamyl-C^{14}-benzene was followed by selenium dioxide oxidation of the *p*-chlorodesoxybenzoin thus formed.

Dibenzoylmethane(1,3-diphenyl-1,3-propane-dione-1-C^{14}). J. D. Roberts, Massachusetts Inst. of Technology

1-(carbonyl-C^{14}-phenyl-)-2-phenyl-1,2-dibromoethane was treated with sodium methoxide and then acidified with hydrochloric acid to give a 49% yield.

Benzoins, benzils, etc., from benzaldehyde-7-C^{14}. O. K. Neville, Oak Ridge Natl. Lab.

A typical reaction sequence is:
4-methoxybenzoin—from anisaldehyde-7-C^{14} (formyl-C^{14}-*p*-methoxybenzene); 65% yield.
4-methoxybenzil—quantitative oxidation by copper sulfate and pyridine.
4-methoxybenzillic acid—alcoholic sodium hydroxide for 6 days; 73% yield.
4-methoxybenzophenone—oxidation by chromic anhydride; 64% yield.

Cyclopentanone-1-C^{14}. R. J. Speer, D. R. Carmody, M. L. Humphries, A. Roberts, Texas Research Foundation

20 mmoles of adipic acid were mixed with 0.25 mmole of barium carbonate-C^{14} in a micro distillation still and heated at 300° C for 2 hours and then at 330° C for an additional hour. The total distillate was collected and dried in ether over anhydrous Na_2SO_4. Upon removal of the ether solvent a 70–80% yield was obtained.

Chrysene-5,6-C^{14}. C. J. Collins, G. Toffel, A. R. Jones, Oak Ridge Natl. Lab.

This polynuclear hydrocarbon was synthesized according to the general method of Collins, *J. Am. Chem. Soc.* **70,** 2418 (1948). Degradation studies confirmed the positions which were labeled.

Cyclopentyl carbinol-C^{14}. R. J. Speer, D. R. Carmody, *et al.*, Texas Research Foundation

The Grignard carbonation of cyclopentylmagnesium bromide yielded the 1-labeled acid which was then reduced by lithium aluminum hydride in chilled ether. Essentially a quantitative yield was obtained.

Methyl-C^{14}-cyclohexane. B. A. Fries, California Research Corporation

This compound was prepared in 75% yield by a Grignard carbonation of bromobenzene followed by hydrogenation of the ethyl ester of benzoic acid using copper chromite catalyst and 200 atm pressure for four hours.

2,4-Dichlorophenoxyacetic acid-1-C^{14}. R. J. Speer, H. Mahler, A. Roberts, *Science* **110,** 562 (1949)

p-Dimethyl-$C_1{}^{14}$-aminoazobenzene. R. A. Boissonnas, R. A. Turner, V. du Vigneaud, *J. Biol. Chem.* **180,** 1053 (1949)

Direct methylation of *p*-aminoazobenzene with methyl iodide-C^{14} led to a mixture of compounds; attempts to modify the reaction were unsuccessful. A one-step synthesis, using methyl iodide-C^{14} and sodium in liquid ammonia, converted monomethylaminoazobenzene to the desired compound. Methyl iodide-C^{14} was bubbled into an equivalent amount of the monomethylaminoazobenzene in liquid ammonia containing dissolved sodium. The yield was 43.5%.

***1,3,5-Trimethylcyclohexane-1,3,5-C^{14}.** A. V. Grosse, S. Weinhouse, *Science* **104,** 402 (1946)

Cyclohexanone-1-C^{14}. R. J. Speer, D. R. Carmody, M. L. Humphries, A. Roberts, Texas Research Foundation

Barium carbonate-C^{14} was heated with a large excess of pimelic acid in a micro distillation tube. The distillate was collected and dried to give a product yield of 60–70%.

This method has proved most satisfactory for the thermal decomposition of carboxylic acids and their salts. Other methods tried included heating the calcium, sodium and lead salts, as well as other barium-containing material, as catalysts.

Cyclohexanone-2-C^{14}; cycloheptanone-2-C^{14}. R. T. Arnold, Univ. of Minnesota, AECU-575

$HC^{14}N$ was passed into an ether solution of a cyclopentanone to give the cyanhydrin which was then hydrogenated over either platinum or Raney-nickel in ethanol containing acetic acid to give the 1-hydroxy-1-methylamino cyclopentane. Upon treatment with nitrous acid, the Tiffeneau rearrangement occurred to produce cyclohexanone-2-C^{14} in 25% yield. Cycloheptanone-2-C^{14} was produced in 45% yield using this same method. It was experimentally shown that the active carbon was adjacent to the carbonyl carbon.

Note: The catalytic hydrogenation led to reductive cleavage of the hydroxyl group when hydrochloric acid was present. Lithium aluminum hydride failed to reduce the cyanohydrin.

***Mesitylene-1,3,5-C^{14}.** A. V. Grosse, S. Weinhouse, *Science* **104,** 402 (1946)

Meso-inositol-$C_6{}^{14}$; scyllitol-$C_6{}^{14}$. R. Anderson, H. Prosser, Brookhaven Natl. Lab.

Potassium carbonyl-C^{14} was prepared by breaking a sealed bulb of potassium in an atmosphere of $C^{14}O$. This material was hydrolyzed by condensing absolute ethanol onto it at −75° C and allowing the mixture to come to room temperature.

For reduction to cyclitols, acetic acid was added to a 40% ethanol solution and hydrogenated over Raney-nickel at 2500 psi. The resultant material yielded uniformly labeled scyllitol and meso-inositol, the latter in 12% yield based on $C^{14}O$.

Naphthalene-1-C^{14}. R. J. Speer, D. R. Carmody, M. L. Humphries, A. Roberts, Texas Research Foundation

This compound was obtained from α-tetralone-1-C^{14} by two procedures. Heating α-tetralone and palladium on charcoal in a sealed tube at 270° C for 5 hours gave a 72% yield of naphthalene-1-C^{14} and 10% yield of α-naphthol-1-C^{14}.

A Clemmensen reduction of α-tetralone to tetralin was followed by heating with Raney-nickel catalyst for 19 hours at ~215° C. This last step gave an 85% yield of crude naphthalene.

Nicotinamide-7-C^{14} (3-carbamyl-C^{14}-pyridine). A. Murray, A. R. Ronzio, Los Alamos Sci. Lab. AECU-87; A. Murray, W. W. Foreman, W. Langham, *J. Am. Chem. Soc.* **70,** 1037 (1948)

Nicotinic acid (3-carboxy-C^{14}-pyridine), prepared according to the second reference given above, was refluxed with thionyl chloride. Later anhydrous ammonia was dis-

tilled into the flask containing the nicotinoyl chloride to give the desired product in excellent yields. The ammonolysis of methyl nicotinate gave only poor yields of the desired 3-carbamyl-C^{14}-pyridine.

***Nicotinic acid-[carboxyl-C^{14}].** A. Murray, W. W. Foreman, W. Langham, *J. Am. Chem. Soc.* **70,** 1037 (1948)

***Phenanthrene-9-C^{14}.** C. J. Collins, *J. Am. Chem. Soc.* **70,** 2418 (1948)

***Phenylacetamide-α-C^{14}.** W. G. Dauben, J. C. Reid, P. E. Yankwich, unpublished experiments (page 196)

***Phenylacetic acid.** W. G. Dauben, J. C. Reid, P. E. Yankwich, unpublished work (page 180); also C. Huggett, R. Arnold, T. Taylor, *J. Am. Chem. Soc.* **64,** 3043 (1942)

Phthalic anhydride-1,2-C_2^{14}. R. F. Nystrom *et al.*, Univ. of Illinois

A Diels-Alder condensation was carried out between maleic anhydride-2,3-C_2^{14} and butadiene in nitrobenzene solvent by heating at 100° C for six hours. A yield of 95% *cis* Δ^4-tetrahydrophthalic anhydride was isolated. The dimethyl ester of Δ^4-tetrahydrophthalic anhydride-1,2-C_2^{14} was reacted with *N*-bromosuccinimide to brominate the two allyl positions adjacent to the double bond. Dehydrohalogenation and saponification yielded phthalic acid-1,2-C_2^{14} from which the anhydride was formed in 80% yield.

Phthalic acid-3-C^{14}. C. J. Collins, Oak Ridge Natl. Lab.

2-methyl-1,4-naphthoquinone-8-C^{14} was oxidized in hot alkaline permanganate. After ether extraction and purification, a 40–50% yield was obtained.

Salicylic acid-7-C^{14} (carboxy-C^{14}-o-hydroxybenzene). P. K. Smith, H. G. Mandel, George Washington Univ. Med. School

The Schmitt modification of the Kolbe synthesis was further modified to suit tracer level work. Instead of an excess of CO_2, a twofold excess of sodium phenolate was heated under pressure with $C^{14}O_2$ at a temperature of 130° C for several hours. The yield was about 65–70%.

Styrene-α-C^{14}; methylphenylcarbinol-1-C^{14}. O. K. Neville, H. E. Heller, J. Weinberger, Oak Ridge Natl. Lab.

Acetyl chloride-1-C^{14} was prepared from sodium acetate-1-C^{14} and used in a Friedel-Crafts condensation with benzene. The resulting aceto-1-C^{14}-phenone was reduced to methylphenylcarbinol-1-C^{14} by lithium aluminum hydride to give an over-all yield of 67%. The benzene solution of crude acetophenone was added dropwise to a stirring solution of $LiAlH_4$ in dry ether. After the reaction was complete, aqueous hydrolysis was followed by ether extraction of the desired product.

Methylphenylcarbinol-1-C^{14} was dehydrated to styrene over potassium bisulfate at 210° C. After careful purification by fractional distillation and chromatography, thermal polymerization of the styrene-α-C^{14} was carried out successfully in a heavy walled Pyrex bomb.

α-Tetralone-1-C^{14}. R. J. Speer, D. R. Carmody, M. L. Humphries, A. Roberts, Texas Research Foundation

γ-phenylbutyric acid-1-C^{14} was synthesized in 65% yield via a Grignard carbonation. The method of Fieser and Hershberg, *J. Am. Chem. Soc.* **61,** 1272 (1939), was used to synthesize α-tetralone in over 90% yield. γ-phenylbutyric acid-1-C^{14} and anhydrous HF were allowed to stand for about 20 hours. The product was ether extracted.

Toluene-1,3,5-C_3^{14}. D. M. Hughes, J. C. Reid, *J. Org. Chem.* **14,** 516 (1949)

Toluene was synthesized in 60% yield by decarboxylating uvitic acid prepared from pyruvic acid-2-C^{14} via alkaline trimerization to methyl dihydrotrimesic acid (67% yield). Oxalic acid-1-C^{14} was obtained as a second product in this synthesis.

1-Phenacyl-1-C^{14}-4-n-amylpyridinium-p-toluene sulfonate. R. G. Pearson, P. Sellers, Northwestern University and Argonne National Laboratory

Carboxy-C^{14}-benzene was converted to benzoyl chloride by thionyl chloride and thence, by diazomethane, to diazoaceto-1-C^{14}-phenone. The latter compound was converted to the quaternary salt in 46% over-all yield (from $BaCO_3$) by treatment with 4-*n*-amylpyridinium tosylate.

5. HETEROCYCLICS

The satisfactory synthesis of labeled guanidine, urea, etc., has provided the impetus for much of the work in the heterocyclic field. Such important compounds as 8-azaguanine and 2,6-diaminopurine have been made available by the Southern Research Institute and are in considerable demand for cancer research. Similarily, uracil and thiouracil are finding wide application in research. Also, for cancer, emphasis is being placed on syntheses of folic acid.

Adenine sulfate-4,6-C_1^{14}. E. Bennett, M. Calvin, Univ. of California Radiation Lab.

4,5,6-Triaminopyrimidine sulfate was heated at 160° C with formamide-formic acid to form adenine in 80% yield after isolation as the sulfate. The over-all yield from cyanide was 20%.

Folic acid (pteroyl-2-C^{14}-glutamic acid). C. T. Fong, C. M. Judson, N. H. Marsh, D. J. Salley, American Cyanamid Company, Stamford, and Lederle Laboratories

Guanidine-hydrochloride-C^{14} was condensed with ethyl cyanoacetate in presence of sodium methoxide to yield diaminohydroxypyrimidine-2-C^{14}. Nitrous acid followed by reduction formed 2,4,5 triamino-6-hydroxypyrimidine in 90% yield based on guanidine.

The product obtained from the reaction of *p*-aminobenzoylglutamic acid with α,α,α'-tribromoacetone was added to a solution of triaminohydroxypyrimidine to form pteroylglutamic acid labeled in the 2 position of the ring. Purification was accomplished by precipitation of the zinc and magnesium salts. Over-all yield was 1% of material of purity greater than 90%.

Folic acid (pteroyl-2-C^{14}-glutamic acid). H. E. Skipper *et al.*, Southern Research Inst.

Guanidine hydrochloride-C^{14} was condensed with ethyl cyanoacetate to form 2,4,5-triamino-6-hydroxypyrimidine-2-C^{14} in a 59% over-all yield from barium carbonate. The pyrimidine hydrochloride solution was swept with nitrogen and an aqueous solution of *p*-aminobenzoyl-1(+)-glutamic acid was added. An absolute ethanol solution of 2,2,3-tribromopropionaldehyde was added very slowly with stirring and the mixture subsequently raised in temperature to 65–70° C. After cooling overnight, the crude mixture was subjected to purification.

Difficulty in achieving satisfactory purification conditions has been experienced.

Guanine hydrochloride-4-C^{14}. E. Bennett, M. Calvin, Univ. of California.

2,4,5-Triamino-6-hydroxypyrimidine sulfate-4-C^{14} was refluxed with 98–100% formic acid and sodium formate; the resulting guanine was isolated as the hydrochloride in 85% yield. The over-all yield from cyanide was 45%.

Guanine-2-C^{14}. H. E. Skipper, L. L. Bennett, Southern Research Inst.

2,4,5-Triamino-6-hydroxypyrimidine-2-C^{14} was reacted with formic acid in which some sodium had been dissolved. After refluxing for 8 hours, the excess formic acid was evaporated and the residue dissolved in sulfuric acid. Guanine was duly isolated and purified in 60–70% yield. Over-all yield from $BaC^{14}O_3$ was 20–25%.

8-Azaguanine-2-C^{14}. H. E. Skipper, L. L. Bennett, Southern Research Inst.

Sodium nitrite was added to an alkaline solution of 2,4,5-triamino-6-hydroxypyrimidine-2-C^{14} after which acetic acid was used to acidify the solution. After heating on a steam bath for one hour and standing overnight in an ice box, 8-azaguanine was isolated and purified in 72% yield. Over-all yield from $BaC^{14}O_3$ was 20–25%.

8-Azaguanine-4-C^{14}. E. Bennett, M. Calvin, Univ. of California Radiation Lab.

2,4,5-Triamino-6-hydroxypyrimidine sulfate-4-C^{14} was dissolved in a sodium acetate solution. sodium nitrite added, and the solution heated for ½ hour. The crude product was isolated by acidification with acetic acid and recrystallized from dilute ammonium hydroxide to yield 8-azaguanine-4-C^{14} in 90% yield. The yield was 45% from cyanide.

7-Amino-1-v-triazolo-pyrimidine-4,6-C_1^{14} (8-aza-adenine-4,6-C_1^{14}). E. Bennett, M. Calvin, Univ. of California Radiation Lab. (*Continued on next page.*)

4,5,6-Triaminopyrimidine sulfate was reacted with nitrous acid in a sodium acetate-acetic acid solution. The product was recrystallized from dilute ammonium hydroxide to yield 7-amino-1-v-triazolo pyrimidine in 80% yield. The over-all yield from cyanide was 20%.

2,4,5,6,-Tetraminopyrimidine-2-C^{14}. H. Skipper, L. Bennett, Southern Research Inst.

Guanidine hydrochloride-C^{14} was condensed in *n*-butanol with phenylazomalononitrile and the azo linkage reduced to the amine by zinc dust to give a 25% yield of product. The condensation of guanidine with malononitrile gave yields of only 13–20%.

2,4,5-Triamino-6-hydroxypyrimidine-2-C^{14}. H. E. Skipper, L. L. Bennett, Southern Research Inst.

Guanidine-C^{14} was prepared from barium carbonate via fusion of the cyanamide with ammonium nitrate. Guanidine hydrochloride-C^{14} was added to sodium in absolute alcohol and after addition of ethyl cyanoacetate, the solution was refluxed in a water bath for 3 hours. To the condensation product, i.e., 2,4-diamino-6-hydroxypyrimidine-2-C^{14}, nitrous acid was added to give the 5-nitroso derivative. Reduction with sodium hydrosulfite gave the desired product. The yield based on guanidine was 60–70%.

2,4,5-Triamino-6-hydroxypyrimidine sulfate-4-C^{14}. E. Bennett, M. Calvin, University of California Radiation Lab.

Ethyl cyanoacetate-3-C^{14} was prepared from sodium cyanide and chloroacetic acid. Guanidine hydrochloride in a sodium methoxide solution was condensed with the ethyl cyanoacetate. The condensation product, 2,4-diamino-6-hydroxypyrimidine, was nitrosated and reduced with sodium hydrosulfite to yield 2,4,5-triamino-6-hydroxypyrimidine sulfate-4-C^{14} in 60% yield from cyanide.

4,5,6-Triaminopyrimidine sulfate-4-C^{14}. E. Bennett, M. Calvin, Univ. of California Radiation Lab.

Phenylazomalononitrile-1-C^{14} was prepared from ethyl cyanoacetate-3-C^{14} through the intermediates, cyanoacetamide-3-C^{14} and malononitrile-1-C^{14}, essentially by the procedure of Cavalieri, *et al.*, *J. Am. Chem. Soc.* **71,** 533 (1949). Modifications in the method increased the over-all yield from sodium cyanide to 50%.

Phenylazomalononitrile was condensed with formamidine hydrochloride in sodium butoxide to yield 4,6-diamino-5-phenylazopyrimidine-4-C^{14}. This was reduced by zinc dust in glacial acetic acid and isolated as the 4,5,6-triaminopyrimidine sulfate-4-C^{14} in 50% yield from phenylazomalononitrile. The over-all yield from cyanide was 25%.

Thiouracil-2-C^{14}. H. E. Skipper, L. L. Bennett, Southern Research Inst.

Using thiourea-C^{14} and the sodium salt of formylacetic ester, a condensation was carried out in water to give a 12% yield of thiouracil. The yield was less in absolute or 95% ethanol using sodium ethylate as catalyst.

A better method appears to be the condensation of thiourea-C^{14} with ethyl β,β-diethoxypropionate in dry ethanol, *n*-propanol or *n*-butanol. Consistent yields of 35–50% of purified thiouracil-2-C^{14} were obtained.

6-Propylthiouracil-2-C^{14}. A. Murray, A. R. Ronzio, Los Alamos Sci. Lab. AECU-621

Thiourea-C^{14} was condensed with ethyl-β-ketocaproate in a solution of absolute ethanol containing sodium metal. After heating for 12 hours at 85° C, the product was isolated and purified to give a 52% yield.

There was some indication that 6-propyl uracil contaminated the product. This arises because of the ready loss of the sulfur of thiourea in basic solution and its replacement by oxygen.

Gramine (3-indolyl-methyl-C^{14}-dimethylamine). C. Heidelberger, *J. Biol. Chem.* **179,** 139 (1949)

Formaldehyde-C^{14} was added to dimethylamine, glacial acetic acid, and indole. After standing for 18 hours, the solution was added dropwise to NaOH to yield crystalline gramine. Yield was 33% (based on CO_2). The formaldehyde was prepared by oxidation of methanol-C^{14} over copper catalyst at 600° C in yields of 40–60%.

3-Indolyl-acetic acid-2-C^{14}. C. Heidelberger, *J. Biol. Chem.* **179,** 139 (1949)

Gramine (3-indolyl-methyl-C^{14}-dimethylamine) and KCN were dissolved in 50% ethanol after which dimethyl sulfate was added and the mixture refluxed. The resultant

nitrile was extracted and refluxed with alcoholic KOH. After evaporation of the alcohol and addition of water, the nitrile was ether extracted and acidified to give indole acetic acid in 50% yield.

Leucopterin-8,9-C_2^{14}; xanthopterin-8,9-C_2^{14}. R. Anker, Western Reserve Univ. Med. School

Oxalic acid-1,2-C_2^{14} was heated with excess 2,5,6-triamino-4-hydroxypyrimidine in a sealed tube at 250° C. Consistent yields of 80% were obtained under carefully controlled conditions. The resultant leucopterin was reduced to xanthopterin by the use of sodium amalgam and ethylene glycol at boiling temperatures. Many by-products were formed and the yield was only 40%. The method of Hitchings, *J. Am. Chem. Soc.* **71,** 741 (1949), failed to yield any xanthopterin on a small scale.

2,6-Diaminopurine-2-C^{14}. H. E. Skipper, L. L. Bennett, Southern Research Inst.; also A. Bendich, J. Tinker, G. Brown, *J. Am. Chem. Soc.* **70,** 3109 (1948).

The formic acid-formamide ring closure on 2,4,5,6-tetraminopyrimidine-2-C^{14} reported by Bendich was found to proceed in 60% yield on a semi-micro scale. 98% formic acid plus anhydrous formamide were used and the entire mixture heated in a sealed tube at 165° C for 3 hours.

Uracil-2-C^{14}. H. E. Skipper, L. L. Bennett, Southern Research Inst.

A direct condensation between urea-C^{14} and ethyl β,β-diethoxypropionate was unsuccessful. The desulfurization of thiouracil-2-C^{14}, however, proceeded in 75–80% yield through the use of chloroacetic acid with refluxing. The over-all yield from $BaC^{14}O_3$ was 11%.

Orotic acid (uracil-4-carboxylic acid-6-C^{14}). D. W. Wilson, Univ. of Pennsylvania

The method of Nye and Mitchell, *J. Am. Chem. Soc.* **69,** 1382 (1948), was used with two modifications. The condensation of aspartic acid-4-C^{14} and potassium cyanate was carried out in a slightly acid solution, *p*H 5.5, rather than in an equivalent amount of alkali. This solution stood at room temperature for 18 hours.

The bromination of 5-carboxymethylidene hydantoin was carried out with equimolar quantities of reagents. The sealed tube required periodic shaking during the 3-hour heating period. The over-all yield of orotic acid, labeled in the 6 position of the pyrimidine ring, was about 40%.

Orotic acid (uracil-4-carboxylic acid-6-C^{14}.) C. Heidelberger, R. Hulbert, Univ. of Wisconsin Med. School

A condensation was carried out in toluene solution between ethyl acetate-1-C^{14} and excess ethyl diethoxyacetate with sodium hydride. The resultant sodium ethyl diethoxyacetoacetate-1-C^{14} was treated directly with thiourea in alcoholic solution. A yield of 69%, based on sodium acetate, of thiouracil-4-aldehyde-6-C^{14} was obtained. The thio group was then converted to the keto group with peroxide and, after hydrolysis, the aldehyde was oxidized to the acid by chromic acid. The over-all yield from barium carbonate-C^{14} was 38%.

Dixanthyl ureide-C^{14}. W. D. Armstrong, Univ. of Minnesota, Dept. of Physiological Chemistry

Urea-C^{14} was used to prepare this compound according to the method of Fosse in "Quantitative Clinical Chemistry," (Methods Volume, page 553, authors: J. Peters, D. Van Slyke). Essentially quantitative yield.

6. STEROIDS

Although an unusual amount of interest has been expressed in this field, the progress toward ring-labeled steroids has been slow. Several groups throughout the country are effectively participating in programs toward this goal.

NOTE: Asterisk (*) before the compound denotes reference appeared in "Isotopic Carbon."

Dehydroandrosterone-16-C^{13}. E. Hershberg *et al.*, *Arch. Biochem.* **19,** 300 (1948)

This is of interest as a route to the C^{14}-containing compound. Through the use of

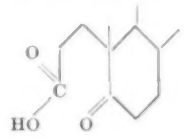
$KC^{14}N$ and by adding the side chain by known methods, cholesterol can be prepared.

***20-Methylcholanthrene-11-C^{14}.** W. G. Dauben, *J. Org. Chem.* **13,** 313 (1948)

***Cholestenone-3-C^{14}.** R. B. Turner, *J. Am. Chem. Soc.* **69,** 726 (1947)

Cholestenone-3-C^{14}; -4-C^{14}. R. Heard, P. Ziegler, McGill Univ., Project (C-876)

This procedure is somewhat similar to Turner's method and involves ozonolysis of cholestenone to 3//5-cholestan-5-one-3-oic acid. (*I*). Its methyl ester was condensed with radioactive bromoacetic acid ester (Reformatsky reaction) and then underwent a Dieckmann cyclization, etc., to yield 14% pure crystalline cholestenone (based on labeled methyl bromoacetate). Three crystalline products were isolated from the Reformatsky step. The detailed study of this reaction course was aided by using both methyl bromoacetate-1 and -2-C^{14}. Intermediates were separated by chromatography. The various reaction products are discussed in detail.

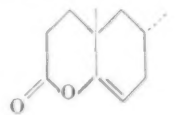


(*I*)

Cholestenone-3-C^{14};-4-C^{14}. R. B. Turner, *J. Am. Chem. Soc.* **72,** 579 (1950)

The ozonization of cholestenone to the keto acid was followed by converting the latter to the enol-lactone by acetic anhydride and acetyl chloride by refluxing for 44 hours. The crude yield was 92%.

The enol-lactone (see diagram) was condensed with phenyl acetate-C^{14} in benzene solution in the presence of sodium hydride. The evolution of hydrogen ceased after 42 hours of stirring under nitrogen and the mixture was acidified with HCl and ether extracted. Hydrolysis, decarboxylation and cyclization were effected simultaneously by treatment with dilute alkali or concentrated HCl in acetic acid. The over-all yield was 40–50% based on the enol-lactone. Phenyl acetate-2-C^{14} yielded cholestenone-4-C^{14} whereas phenyl acetate-1-C^{14} yielded cholestenone-3-C^{14}.



Cholesterol-T. D. Kritchevsky, Univ. of California Radiation Lab.

This compound was prepared by shaking cholesterol with reduced platinum oxide, glacial acetic acid, and tritium-enriched water.

Cholesterol-6-C^{14}. F. N. Hayes, Illinois Inst. of Technology

Cholesterol was oxidized to the 7-keto derivative and condensed with methyl-C^{14}-magnesium iodide to give the 7-methyl-C^{14}-7-hydroxy compound. The use of osmium tetroxide resulted in hydroxylation of the 5,6 double bond. The 6-carbon was split out when glycol cleavage was accomplished by lead tetracetate. A base-catalyzed condensation reclosed the ring using the former 7-methyl-C^{14} carbon as the new 6-ring carbon of the 7-keto cholesterol. Reduction then afforded cholesterol in good yield. Preliminary work on this sequence of reactions appears promising.

Estrone-16-C^{14}. R. Heard, J. Saffran, L. Thompson, McGill Univ., Project (C-876)

Ring D of estrone methyl ether was opened to give a mixture of the dimethyl ester and the half ester of marrianolic acid methyl ether. After mild saponification, the half ester was converted to its acid chloride and treated with radioactive diazomethane. The crude mixture was subjected to the Arndt-Eistert rearrangement with silver oxide in methanol to yield the dimethyl ester of homo-marrianolic methyl ether. Since the Dieckmann cyclization gave low yields, the ester was converted to the lead salt which cyclized at 270° C in 65% yield. The over-all yield of estrone was 15% on a semi-micro scale. Hot runs are now being started.

Hexestrol 2,3,4,5-T. D. L. Williams, A. R. Ronzio, Los Alamos Sci. Lab. AECU-714

Both meso- and racemic forms of hexestrol, labeled with tritium, were obtained by the hydrogenation of dienestrol with a palladium-on-carbon catalyst. The presence of a small amount of tritium during hydrogenation consistently exerted a significant influence upon the rate of hydrogenation. A yield of 95% was obtained.

Methyl ester 3-keto-Δ^4-etiocholenic acid-4-C^{14}. R. Heard, P. Ziegler, McGill Univ., Project (C-876)

This compound was synthesized by the same series of reactions as employed for

labeling cholestenone. Ozonolysis was followed by condensation of the methyl ester with bromoacetic ester-2-C^{14}. The Dieckmann condensation reformed the ring to give a 28% over-all yield of the desired compound.

Progesterone-21-C^{14}. B. Riegel, F. S. Prout, *J. Org. Chem.* **13,** 933 (1948)

This was prepared by the alkylation of 3-keto-4-etiocholenoyl chloride with di(methyl-C^{14})-cadmium.

Methyl-C^{14}-magnesium Grignard was converted to di(methyl-C^{14})-cadmium by the usual procedure of adding anhydrous cadmium chloride. Progesterone was separated and purified by chromatographic techniques and molecular distillation.

Progesterone-21-C^{14}. R. D. Heard, C. Yates, McGill Univ., Montreal

Di(methyl-C^{14})-cadmium was reacted with 3-keto-4-etiocholenic acid chloride to give progesterone in 25% yield. This method may not be completely reliable. Although progesterone was usually the main product, one run gave a main product which was the methyl ester of the starting acid, i.e., 3-keto-4-etiocholenic methyl ester. Both this compound and progesterone exhibit approximately the same ultraviolet absorption spectrum, melting point (with only a slight depression on admixture), and optical rotation. In five early experiments, the data were insufficient to establish positive identification but indicated progesterone in each case.

Progesterone-21-C^{14}. H. B. MacPhillamy, C. R. Scholz, *J. Biol. Chem.* **178,** 37 (1949)

The cadmium Grignard of methyl-C^{14}-iodide was reacted with 3-acetoxy-5-etio cholenyl chloride to yield 50% 21-C^{14}-Δ^5-pregnene-3-ol-20-one acetate. Saponification and oxidation by the Oppenauer method, using aluminum isopropylate, yielded progesterone-21-C^{14}. Over-all yield was 26% based on methyl-C^{14}-iodide.

Stilbestrol [p,p'-dihydroxy-di(ethyl-2-C^{14})stilbene]. E. F. Schoenwaldt, G. H. Twombly, Columbia Univ. College of Physicians and Surgeons

This compound was prepared by the Grignard addition of ethyl iodide-2-C^{14} to α-ethyl desoxyanisoin. The resultant carbinol was dehydrated with potassium acid sulfate and then ether cleavage with KOH in diethylene glycol yielded 27% (over-all) stilbestrol. A twofold excess of Grignard reagent over α-ethyl desoxyanisoin gave a 96% yield (based on ketone). An equimolar mixture gave an oily product difficult to handle.

The use of *p*-toluene sulfonic acid as a dehydrating agent was unsatisfactory and gave poor yields. Potassium acid sulfate, however, gave a 52% yield.

17-Methyl-C^{14}-testosterone. H. MacPhillamy, C. Scholz, *J. Biol. Chem.* **178,** 37 (1949)

Methyl-C^{14} iodide was employed as a Grignard reagent on androstene-3,17-dione-3-enol to give a one-step yield of 42% for 17-methyl-C^{14}-testosterone. Extraction and chromatography were employed in its purification.

***Testosterone-3-C^{14}.** R. B. Turner, *Science* **106,** 248 (1947)

Testosterone-3-C^{14}; -4-C^{14}. R. B. Turner, *J. Am. Chem. Soc.* **72,** 579 (1950)

Condensation of the enol-lactone derived from testosterone benzoate with phenyl acetate-C^{14} was effected by essentially the same procedure as was used for the synthesis of cholestenone. Phenyl acetate-2-C^{14} yielded the 4-labeled compound while phenyl acetate-1-C^{14} yielded testosterone-3-C^{14} in about 50% yield based on the enol-lactone.

7. VITAMINS

Here again the demand for labeled vitamins is large whereas the progress is slow. The synthesis of vitamin K illustrates the great potentialities of chemical synthesis in labeling some of these characteristically complex materials. There is a great need for more workers to enter this important field.

Thiamin (4-methyl-5-hydroxyethyl-thiazole-2-C^{14}). D. L. Williams, A. R. Ronzio, Los Alamos Sci. Lab.

Thiourea-C^{14} was condensed with 3-aceto-3-chloropropanol in 84% yield to form the

substituted thiazole. The deamination of 2-amino-4-methyl-5-hydroxyethyl-thiazole-2-C^{14} was accomplished by diazotization and reduction with H_3PO_2 in 6*N* hydrochloric acid. The yield in this step was about 60–70%.

The thiazole was then condensed with 2-methyl-4-amino-5-methyl-pyrimidine hydrobromide by adding the latter compound dropwise. In butyl alcohol as solvent, some difficulty was experienced in that the butyl ester of the pyrimidine was formed to some extent and part of the thiazole was consumed to form the ether of the radioactive thiamin. Thiamin was isolated as the picrate and the over-all yield was approximately 25%.

Vitamin C [l-ascorbic acid-1-C^{14}]. J. L. Burns, C. G. King, *Science* **111,** 257 (1950)

The addition of radioactive cyanide to l-xylosone yielded imino-l-ascorbic acid which, upon hydrolysis, produced ascorbic acid-1-C^{14}. The crystalline product was obtained in 25% yield after purification by ion-exchange techniques.

The cyanide addition was conducted at the optimum *p*H of 9.4–10.4, the xylosone being present in slight excess. The excess was consumed later with inert sodium cyanide since it would have interfered with the crystallization of ascorbic acid. Some decarboxylation of ascorbic acid occurred during the hydrolysis step.

Vitamin K [2-methyl-1,4-naphthoquinone-8-C^{14}]. C. J. Collins, Oak Ridge Natl. Lab.

Inactive 3-tolyl-propylbromide was prepared via reduction of *p*-methyl cinnamic acid and subjected to a Grignard carbonation with $C^{14}O_2$. Yield was 85%.

The acid chloride was prepared using PCl_5 and the subsequent addition of stannic chloride led to ring closure to give 7-methyl-1,2,3,4-tetrahydronaphthenone-1-C^{14} in about 90% yield.

Reduction over palladium-on-charcoal catalyst in glacial acetic and perchloric acids gave essentially a quantitative yield of 7-methyl-1,2,3,4-tetrahydronaphthalene-1-C^{14}. Dehydrogenation at 220° C over palladium-on-charcoal catalyst for 22 hours produced 2-methylnaphthalene-8-C^{14}.

Chromic oxide in glacial acetic acid resulted in oxidation to 2-methyl-1,4-naphthoquinone-8-C^{14}. Yield of purified material was 52%.

8. SUGARS

By far the most popular method for the synthesis of 1-labeled sugars is the method of Sowden and Fischer which employs a condensation with labeled nitromethane.

The demand for labeled sugars of various types has been quite large and, at the present time, a limited amount of work is being initiated in the hopes of remedying this situation.

d-Glucose-1-C^{14}. H. A. Mahler, Univ. of California Radiation Lab. AECD-2400 (Sept., 1948)

This is a detailed thesis covering such related topics as the synthesis of nitromethane-C^{14}, the cyanohydrin reaction, and the Nef reaction.

The condensation of arabinose with nitromethane-C^{14} was investigated. The sodium salts of 1-desoxy-1-nitro-d-glucose and d-mannose were converted, in 95% yield, to the corresponding free sugar nitro alcohols by ion exchange with either Dowex-50 or Amberlite IR-100 H. The Nef reaction was carried out at −15° C using 17*N* H_2SO_4 to convert the nitro alcohols into d-glucose and d-mannose in about 65% yield.

Over-all yields, based on methyl iodide-C^{14}, were 7% for d-mannose-1-C^{14} and 5% for d-glucose-1-C^{14}.

d-Glucose-1-C^{14}; d-mannose-1-C^{14}. J. C. Sowden, *Science* **109,** 229 (1949)

Methyl iodide-C^{14} was reacted with silver nitrite according to the Victor Meyer reaction to obtain nitromethane-C^{14} in good yield.

The condensation of nitromethane with d-arabinose yielded 1-nitro-1-desoxy-d-mannitol and 1-nitro-1-desoxy-d-sorbitol.

The nitroalcohols were converted to the corresponding hexoses by the Nef reaction. Over-all yields are about 10%.

d-Glucose-1-C^{14}; d-mannose-1-C^{14}. H. G. Wood, Western Reserve Univ. Med. School

d-Glucose-1-C^{14} was synthesized in 6–7% yield according to the method of Sowden and Fischer, using nitromethane-C^{14}. d-Mannose-1-C^{14} was obtained in 12–15% yield based on barium carbonate. One modification consisted of replacing the absolute methanol with absolute ethanol after precipitation of unreacted d-arabinose.

9. SEDATIVES AND MISCELLANEOUS COMPOUNDS

The great interest in the use of labeled compounds for medical and physiological research has led to the recent synthesis of a number of important compounds such as codeine, demerol, nembutal, choline, etc. Interest is continuing and considerable synthetic work should appear next year.

NOTE: Asterisk (*) before the compound denotes reference appeared in "Isotopic Carbon."

Chloretone-[1,1,1-trichloro-2-methyl-C^{14}-2-propanol]. E. H. Mosbach, S. S. Jackel, C. G. King, Columbia Univ. Dept. of Chemistry

Low yields precluded the use of direct condensation of acetone and chloroform in the presence of KOH. A Grignard addition of methyl-C^{14} magnesium iodide to a 15-fold excess of ethyl trichloroacetate gave an 11% yield of pure, sublimed product.

Codeine [3-methoxy-C^{14}-morphine]. B. M. Tolbert, *et al.*, Univ. of California Radiation Lab., UCRL-428

Morphine-*N*-oxide was prepared as a starting material to prevent morphine's tertiary amino group from undergoing methylation. Methyl iodide-C^{14} was distilled into a solution of morphine-*N*-oxide in sodium methoxide. After 4 hours of refluxing, the codeine-*N*-oxide was reduced with SO_2 and the desired product extracted. A yield of 60%, based on methyl iodide-C^{14}, was obtained.

Demerol-[N-methyl-C^{14}-4-phenyl-4-carbethoxypiperidine]. W. Tarpey, Univ. of California Radiation Lab.; also H. Hauptmann, B. M. Tolbert, *J. Am. Chem. Soc.* (in press)

Using the Wallach reaction, the alkylation of 4-phenyl-4-carbethoxypiperidine was carried out with formaldehyde-C^{14} and inactive formic acid. Only the demerol formed and the recovered formaldehyde were radioactive. Reduction is effected only by the formic acid and there is no exchange between formic acid and formaldehyde. The yield was about 40%.

Methadone [4,4-diphenyl-6-dimethylamino-3-heptanone-1-C^{14}; -2-C^{14}]. B. M. Tolbert, F. Christenson, F. N. Chang, P. P. Sah, *J. Org. Chem.* **15,** 525 (1949)

Ethylacetate-1-C^{14} was hydrogenated to ethyl alcohol-1-C^{14} over copper chromite catalyst. Ethylmagnesium bromide was then condensed with 2,2-diphenyl-4-dimethylamino pentanenitrile to give a 9.5% over-all yield, based on $C^{14}O_2$, of the 2-labeled compound. Sodium acetate-2-C^{14} was employed for the 1-labeled compound.

Nembutal [pentobarbital-2-C^{14}]. A. Murray, A. R. Ronzio, Los Alamos Sci. Lab., AECU-86

Urea-C^{14} was condensed with ethyl-2-amyl-malonic ester in a sodium ethoxide-alcohol solution by heating in a sealed tube at 85° C for 5 days. This barbiturate was finally ether extracted and gave a yield of crude product of 77%. It was converted to the sodium salt by heating with bicarbonate. Average yields were 70%.

MISCELLANEOUS CARBON-14 COMPOUNDS

Biotin-(ureido-C^{14}). D. B. Melville, J. G. Pierce, C. W. Partridge, *J. Biol. Chem.* **180,** 299 (1949); † P. Melville *et al.*, *Science* **94,** 308 (1941)

Carbon dioxide-C^{14} was incorporated into biotin as the ureido carbon by utilizing the reaction of phosgene with the diamino-carboxylic acid derived from biotin†.

***Ethyl carbamate (urethane).** H. E. Skipper, C. E. Bryan, O. S. Hutchinson, "Techniques for the Synthesis and Metabolic Study of Urethane," Atomic Energy Commission, Isotopes Division Cir. C-8 (Sept., 1947)

Choline [(β-hydroxyethyl)-trimethyl-C_1^{14} ammonium hydroxide]. R. J. Winzler, Univ. of Southern California Med. School

This synthesis was accomplished by the condensation of methyl iodide-C^{14} with 2-dimethylaminoethanol in ether solvent in a closed vessel.

Choline chloride [(β-hydroxyethyl)-trimethyl-C_1^{14} ammonium chloride]; acetyl choline chloride [(β-acetoxyethyl)-trimethyl-C_1^{14} ammonium chloride]. F. Christenson, B. M. Tolbert, Univ. of California Radiation Lab., AECU-373 (July, 1949)

Methyl iodide-C^{14} was used to quaternize 2-dimethylamino ethyl alcohol by heating the mixture in a closed tube at 60° C for $1\frac{1}{2}$ hours. The non-volatile residue was heated with silver oxide to obtain the quaternary base and, after filtration, HCl was added to give choline chloride which was purified to give a yield of 87%.

Acetyl choline chloride was heated in a sealed tube with acetic anhydride for two hours at 100° C. The desired product was crystallized from dry acetone in good yield.

***1,2,5,6-Dibenzanthracene-9-C^{14}.** C. Heidelberger, P. Brewer, W. G. Dauben, *J. Am. Chem. Soc.* **69,** 1389 (1947)

2-Acetylaminofluorene-9-C^{14}. F. E. Ray, C. Geiser, *Science* **109,** 200 (1949)

This carcinogenic compound was synthesized from 2-iodobiphenyl through a Grignard carbonation with $C^{14}O_2$. Ring closure to fluorenone-9-C^{14} was accomplished in 88% yield after which reduction yielded fluorene. Nitration, reduction, and acetylation netted an over-all 20% yield of the final product.

***9-Fluorenecarboxylic acid (carboxyl C^{14}).** C. J. Collins, *J. Am. Chem. Soc.* **70,** 2418 (1948)

***Benadryl hydrochloride.** R. W. Fleming, G. Rieveschl, Jr., abstract of paper presented before Am. Chem. Soc., New York (Sept., 1947)

Methyl-C^{14}-di-(β-chloroethyl)-amine; hydrochloride (nitrogen mustard). W. W. Foreman, A. Murray, A. R. Ronzio, Los Alamos Sci. Lab. AECU-360

Methyl bromide-C^{14} was condensed with diethanolamine in absolute ethanol. (Methyl iodide was found to be decomposed by diethanolamine). When equimolar ratios of methyl bromide and amine were used, 20–35% of the amine was converted to the quaternary salt. A mole ratio of halide to amine of 1 to 5 gave no amine higher than the tertiary product desired. Thionyl chloride was used to halogenate the mixture, after which careful purification procedures gave a 52% yield of the desired product.

Dibromo-Br^{82}-procaine hydrochloride. F. Howarth, *Nature* **161,** 857 (1948); **163,** 679 (1949)

Procaine hydrochloride (C^{14}). G. Mandel, George Washington Univ. Med. School

Stilbene-4,4′-dicarboxamidine-C^{14}-diisethionate. Univ. of California Radiation Lab., J. C. Reid, AECU-586 (Sept., 1949)

This compound was prepared from sodium cyanide-C^{14} to give C^{14} in the amidine groups. Cuprous cyanide was prepared and used to convert 4,4′-dibromostilbene to the 4,4′-dicyano-C^{14} compound in 100% yield. The latter compound was converted to the imido-ester by a mixture of dry HCl in chloroform and methanol. Yield was 79%.

The imido-ester was heated at 60° C for 36 hours in a solution of anhydrous ammonia and ethanol to produce the diamidine (free base). Treatment with free isethionic acid then completed the reaction sequence to give an over-all yield from sodium cyanide-C^{14} of 19%.

10. COMPOUNDS LABELED WITH IODINE-131

Although iodine is being used to label insecticides and medicinals such as penicillin-X, perhaps the most striking application has been in the iodination of the phthalein dyes which are used in the study of tumor localization.

2-Iodo I^{131}-3-nitrobenzoic acid. J. W. Mitchell, J. W. Wood, W. C. Wolfe, G. W. Irving, *Science* **106,** 395 (1947)

This compound was synthesized by diazotization of 3-nitroanthranilic acid followed by treatment with KI^{131} via a Sandmeyer type reaction.

Iodo-I^{131}-casein. C. F. Hamilton, M. H. Power, A. Albert, *J. Biol. Chem.* **178,** 213 (1949)

Iodine crystals containing I^{131} were prepared and added to a solution of casein in a bicarbonate solution containing some colloidal oxides of manganese. At *p*H 4.6, the mixture was incubated. Of the total radioactive iodine, 37% occurred as thyroxine.

Iodo-I^{131} dyes for cancer research. H. Bloch, F. Ray, *J. Natl. Cancer Inst.* **7,** 61 (1946)

The following radioactive materials were synthesized: monoiodo-I^{131}-sulfanilamide, monoiodo-I^{131}-sulfapyridine, iodo-I^{131}-trypan blue. Iodination of various dyes is noted.

Diiodo-I^{131}-fluorescein. G. Boyack, G. E. Moore, D. F. Clausen, NUCLEONICS **3,** No. 4, 62 (1948)

Purified fluorescein was iodinated with iodine chloride-I^{131} in acetic acid. One hour of heating on a steam bath was followed by precipitation of diiodo-I^{131}-fluorescein in distilled water. The yield was 80%.

3,5-Diiodo-I^{131}-folic acid (diiodopteroylglutamic acid). H. E. Skipper, L. L. Bennett, P. E. Edwards, J. A. Johnson, Southern Research Inst.

One millimole of folic acid and two millimoles of sodium bicarbonate were heated in solution to 75° C. To this solution, three millimoles of iodine monochloride-I^{131} were added. After standing for a short time, a trace of sodium bisulfate was added, followed by NH_4OH. The subsequent addition of HCl led to the formation of a precipitate which, after purification, corresponded to a yield of 65% of the product based on iodine.

Diethyl-β-iodo-I^{131}-ethylamine hydrochloride (a nitrogen mustard). A. M. Seligman, A. M. Rutenberg, O. M. Friedman, *J. Natl. Cancer Inst.* **9,** 261 (1949)

This compound was synthesized from diethyl-β-hydroxyethylamine and thionyl chloride in chloroform solution. The free base was formed and treated with radioactive sodium iodide in acetone. The final purified hydrochloride salt corresponded to 70% yield based on sodium iodide.

3′,5′-Diiodo-I^{131}-A-methopterin. H. E. Skipper, L. L. Bennett, P. E. Edwards, J. A. Johnson. Southern Research Inst.

To a hot (75° C) bicarbonate solution containing 2 millimoles of A-methopterin, five millimoles of iodine monochloride-I^{131} were added. During this addition, unchanged A-methopterin partially precipitated. The suspension was then decanted and processed to give a 53% yield based on the iodine content.

Pipsyl chloride-I^{131} (p-iodo-I^{131}-phenyl sulfonyl chloride). M. Levy, A. S. Keston, New York Univ. College of Med.

The method of Keston, Udenfriend, and Cannon, *J. Am. Chem. Soc.* **71,** 249 (1949), has been modified with success.

The sulfite contained in fission iodine-I^{131} must be removed since it interferes with the subsequent reaction with *p*-diazobenzene sulfonic acid. After neutralization, the product was isolated by adding carrier sodium-*p*-iodophenyl sulfonate. The synthesis of the S^{35}-labeled compound is being studied.

Iodo-I^{131}-penicillin-X. D. L. Tabern, G. Gleason, D. Dunnigan, Abbott Laboratories

In an attempt to obtain monoiodo-I^{131}-penicillin, iodination was carried out in bicarbonate solution at 0° C using iodine monochloride-I^{131}. Filter paper chromatographic separations indicated bands corresponding to mono and diiodo-I^{131}-penicillin plus an unidentified band.

Tetraiodo-I^{131}-phenolphthalein. D. L. Tabern *et al.*, Abbott Laboratories

The method used by Moore to prepare iodofluorescein was first tried using KI^{131} and dichloramine-T in acetic acid. It was shown that dichloramine-T caused some decomposition of the phenolphthalein.

The successful procedure involved the addition of iodine monochloride-I^{131} to a dilute aqueous HCl solution of sodium phenolphthalein. It was subsequently found that

better yields were obtained by using glacial acetic acid. Stirring, after mixing, continued for ½ hour at room temperature, 1 hour at 40° C, 1 hour at 55° C. Yield was about 95%.

2,4-dichloro-5-iodo-I^{131}-phenoxyacetic acid (and derivatives). W. C. Wolfe, *et al.*, *J. Org. Chem.* **14,** 900 (1949)

Nitration of 2,4-dichlorophenoxyacetic acid furnished the 5-nitro compound which was reduced by ferrous sulfate and ammonia to 5-amino-2,4-dichlorophenoxyacetic acid. The sodium salt of this latter compound was diazotized in sulfuric acid. Potassium iodide-I^{131} was added and, after purification of the final product, inert carrier was added. Yield was 47% (based on radioactivity).

2,4-dichloro-5-iodo-I^{131}-phenoxyacetic acid. R. J. Speer, H. Mahler, A. Roberts, *Science* **110,** 562 (1949)

Iodotetrazolium salt (2,5-diphenyl-p-iodo-I^{131}-phenyltetrazolium chloride). A. M. Seligman, R. Gofstein, A. M. Rutenberg, *Cancer Research* **9,** 366 (1949)

Aniline was iodinated in bicarbonate solution and then diazotized. The diazonium salt was reacted with benzalphenylhydrazone and, after the mixture was cooled in ice, distilled water was added slowly to precipitate the red pigment formazan. After purification, the formazan was oxidized to the tetrazolium salt by the addition of amyl nitrite and hydrochloric acid.

Thyroxine-I^{131}. E. Frieden, M. B. Lipsett, R. J. Winzler, *Science* **107,** 353 (1948)

This material was prepared by exchange of iodine atoms between radioactive iodide in aqueous butanol and d,l-thyroxine. Up to 30% of the active iodine could be recovered in recrystallized thyroxine.

Monoiodo-I^{131}-monohydroxytrypan blue. C. D. Stevens, A. Lee, *et al.*, *Cancer Research* **9,** 139 (1949)

Radioactive sodium iodide was reacted with diazotized trypan blue by use of the Sandmeyer reaction. The final product was 52% dihydroxy trypan blue, 40% monoiodo-I^{131}-monohydroxy trypan blue and 3% diiodo-I^{131}-trypan blue.

11. COMPOUNDS LABELED WITH SULFUR-35

The two principal fields of interest with regard to this radioisotope are the sulfur-containing amino acids and the organic sulfonic acids of pharmaceutical interest. Considerable success has been experienced in both fields, although these programs are still in their early stages of development.

Cysteine-S^{35}. G. Gleason, D. L. Tabern, Abbott Laboratories

The method of Woods and Van Middlesworth was used with modifications. Serine methyl ester hydrochloride was converted by PCl_5 and acid hydrolysis to α-amino-β-chloropropionic acid. The α-amino-β-chloropropionic acid obtained was condensed with benzylthiouronium-S^{35}-chloride instead of benzyl mercaptan-S^{35}.

Benzylthiouronium chloride-S^{35} was prepared pure in 76% yield from thiourea-S^{35}. The condensation product, S-benzyl cysteine, was formed in 63% yield and reduced with sodium in liquid ammonia, following which oxidation was accomplished by iodine rather than by air. This oxidation method is not completely satisfactory.

The $POCl_3$ must be thoroughly washed out of the α-amino-β-chloropropionic acid to permit satisfactory isolation. Due to the instability of a number of intermediates, the entire set of reactions must be carried out rapidly.

Cystine-S^{35}. E. M. Fry, *J. Org. Chem.* **15,** 483 (1950)

The routes to cystine described in this paper all depend on the initial transformation of serine into 2-phenyl-4-carboxymethyloxazoline. In the first route the hydrochloride salt of the oxazoline was rearranged to methyl-α-benzamido-β-chloropropionate and the chlorine replaced directly by the unprotected sulfhydryl group.

The other useful method is based on the rearrangement of the thiobenzoic acid salt of the oxazoline acid to give N,S-dibenzolcystine.

S-benzyl cysteine-S^{35}; l-cystine-S^{35}. J. L. Wood, L. Van Middlesworth, *J. Biol. Chem.* **179,** 529 (1949)

Elemental sulfur in xylene was prepared by oxidizing radioactive sulfide ion with iodine in KI. Benzylmagnesium chloride was added to the cold solution which was then allowed to stand. After washing with petroleum ether, the solution was made alkaline and stirred in a nitrogen-swept atmosphere. α-Amino-β-chloropropionic acid hydrochloride was added at 50° C to yield S-benzyl-d,l-cysteine-S^{35} after acidification and isolation. Yield was 40%.

l-cystine-S^{35}: S-benzyl-l-cysteine-S^{35} was treated with sodium in dry liquid ammonia. The product was oxidized with air to give a 77% yield of l-cystine-S^{35}.

Methionine-S^{35}. D. L. Tabern, G. Gleason, D. Dunnigan, Abbott Laboratories; H. Tarver, C. Schmidt, *J. Biol. Chem.* **130,** 67 (1939); W. Patterson, V. du Vigneaud, *J. Biol. Chem.* **111,** 393 (1935); H. Snyder, G. Cannon, *J. Am. Chem. Soc.* **66,** 511 (1944)

The process of Tarver and Schmidt gave only 20% yield. The method of Patterson and du Vigneaud gave better yields but involved benzyl mercaptan which is hard to handle. Since thiourea-S^{35} had been synthesized in good yield, the method of Snyder was used with modifications.

Thiourea was condensed with dichlorodiethyldiketopiperazine using equivalent amounts in absolute alcohol. Slow continuous stirring at temperatures not exceeding 110° C took place for 40–48 hours. In the hydrolysis-methylation step, a slow addition of NaOH improved the yield.

Potassium thiocyanate-S^{35}. D. L. Tabern, G. Gleason, D. Dunnigan, Abbott Laboratories

A 0.2*N* iodine solution which was 2*N* in HCl was used to oxidize sulfide-S^{35} to elemental sulfur. Excess iodine was reduced with stannous chloride, and the mixture was allowed to stand overnight. After centrifuging and washing, a few ml of xylene were added and, on heating, complete solution was achieved.

This xylene solution was added to an acetone solution containing more elemental sulfur and potassium cyanate, and the mixture was refluxed for two hours. After filtration, the acetone was evaporated, and the product was washed with ether.

An aqueous solution was boiled for 30 minutes and purged with CO_2 to remove traces of H_2S and HCN. The final crystallization gave a 49% yield of pure product as shown by paper chromatograms. This is a modification of a method described by Wood, *J. Biol. Chem.* **170,** 251 (1947).

Sulfanilamide-S^{35}. J. E. Christian *et al.*, Purdue Univ. School of Pharmacy

Acetic anhydride was added to chilled sulfuric acid containing S^{35} and subsequently acetanilide was added and the mixture heated for 30 minutes on a steam bath.

N-acetylsulfanilic acid-S^{35} was reacted with PCl_5 to yield sulfonyl chloride-S^{35} after which 28% NH_4OH was added and the mixture warmed. After neutralization and purification, the sulfanilamide was obtained in 35% yield.

N-acetylsulfanilyl-S^{35} chloride. I. M. Klotz, J. B. Melchior, *Arch. Biochem.* **21,** 35 (1949)

Since many of the sulfonamides are synthesized from N-acetylsulfanilyl chloride and the appropriate amine, this compound was prepared. To sulfuric acid-S^{35}, cold acetanilide and acetic anhydride were added. The usual method of reacting chlorosulfonic acid with acetanilide was discarded because of low yields. The resultant N-acetylsulfanilic acid was suspended in chloroform and treated with PCl_5 to give the desired chloride.

Sulfanilamide-S^{35} was then prepared in 30% over-all yield.

Tetraethylthiouram disulfide-S^{35}. L. Eldjarn, *Acta Chem. Scand.* **3,** 644 (1949)

Potassium sulfide-S^{35} was prepared from calcium sulfate-S^{35} by reduction with CO at 850° C and conversion to the potassium salt. Carbon disulfide-S^{35} was prepared by allowing exchange to occur between potassium sulfide-S^{35} and CS_2.

The resultant CS_2^{35} was then reacted with diethylamine and potassium hydroxide to give the diethyl carbamate which was oxidized with sodium tetrathionate to yield the desired product.

p-Toluenesulfonyl-S^{35} chloride. L. Soffer, F. E. Ray, Univ. of Cincinnati

By means of sulfuric acid-S^{35} and toluene, the sulfonic acid was obtained in 60% yield. Using PCl_5 and $POCl_3$, the sulfonyl-S^{35} chloride was obtained in an over-all yield of 47% based on H_2SO_4.

2-(p-Aminophenyl)-thiazol-S^{35}. A. Erlenmeyer, H. Meyer, H. Noll, E. Sorkin, *Helv. Chim. Acta* **32,** 1209 (1949)

Thiourea-S^{35}. C. W. Bills, A. R. Ronzio, Los Alamos Sci. Labs. AECU-619; R. V. Heuser—U. S. Patent 1,991,852 (1935)

The procedure of Heuser was modified to utilize all of the radioactive hydrogen sulfide present. Into cyanamide in weak NH_4OH, H_2S^{35} was passed, the entire mixture being frozen out with liquid nitrogen. After heating to 40° C for 24 hours in a sealed flask, vacuum sublimation yielded the desired product in 92% yield.

12. COMPOUNDS LABELED WITH PHOSPHORUS-32

Only a limited amount has appeared in this field and few laboratories are engaged in the chemical synthesis of organic phosphorus compounds.

Diisopropyl fluorophosphate-P^{32}. B. Witten, J. I. Miller, *J. Am. Chem. Soc.* **70,** 3886 (1948)

Dicalcium phosphate was formed from potassium dihydrogen phosphate-P^{32} and heated at 500° C to convert it to the pyrophosphate. This latter material was chlorinated at 700° C to form a mixture of phosphorus pentachloride and the oxychloride. Powdered antimony resulted in reduction of PCl_5 to PCl_3, while powdered charcoal heated in a quartz tube reduced $POCl_3$ to PCl_3.

This PCl_3 was then reacted with isopropyl alcohol and the diisopropyl phosphite-P^{32} was chlorinated and finally converted to the fluoro-compound with anhydrous sodium fluoride. Over-all yield was 33%.

Diisopropyl fluorophosphate-P^{32}. B. C. Saunders, T. S. Worthy, *Nature* **163,** 797 (1949)

Phosphorus containing P^{32} was converted to the trichloride on a small scale and, upon reaction with isopropyl alcohol, was converted to diisopropyl phosphite. Chlorination resulted in the diisopropyl chlorophosphate which, upon treatment with sodium fluoride, yields the desired fluoro-compound.

Diisopropyl fluorophosphate-P^{32}. B. Jandorf, P. McNamara, *J. Pharmacol. Exptl. Therapy* **98,** 77 (1950)

Glycerophosphate-P^{32}. J. W. Spinks, C. C. Lee, J. B. O'Neil, *Can. J. Research* **B27,** 629 (1949)

Sodium dihydrogen phosphate-P^{32} was heated with glycerol under reduced pressure at 180° C for two hours. The glycerophosphate was recovered in 65% yield. It was concluded that this product was a mixture of the α and β sodium glycerophosphates.

p-Nitrophenyl phosphate-P^{32}. B. Axelrod, *J. Biol. Chem.* **176,** 295 (1948)

85% H_3PO_4 containing P^{32} was heated and evaporated until it was anhydrous and then it was mixed with PCl_5 and reheated to give an 80% yield of redistilled $P^{32}OCl_3$. p-Nitrophenol was dissolved in $CHCl_3$ and to this solution was added, in succession, the $P^{32}OCl_3$ and some dry pyridine. Adequate stirring and cooling was necessary. Several pieces of ice were added after 30 minutes. After several hours' standing, the *p*-nitrophenyl phosphoric acid-P^{32} was obtained by chloroform extraction.

Alkyl diphenylphosphine-P^{32} oxides. D. C. Morrison, Univ. of California Radiation Lab., UCRL-594 (Feb. 1950)

This consists of a study of the action of Grignard reagents on the chlorides of aromatic phosphinic acids.

To a solution of the diphenylphosphinic acid-P^{32} in toluene, thionyl chloride was added. After formation of the acid chloride, it was diluted with benzene and

added dropwise to a large excess of Grignard reagent at room temperature. After refluxing for 2 hours, it was cooled, hydrolyzed, and ether extracted to yield the expected product. No reaction occurred between phosphinic acid itself and the Grignard reagent.

Syntheses Being Investigated Under Isotopes Division Contract:

Methods for synthesizing radioactive-labeled compounds will be developed by 11 organizations under contracts awarded for 1950 and 1951 by the Atomic Energy Commission. The organizations are listed below, together with the different compounds being investigated.

Texas Research Foundation, Renner, Texas: Deuteropolyethylene, ring-labeled benzene (high specific activity), C^{14}-labeled ketones from carboxylic acids, and labeled growth regulators (indole acetic acid, naphthalene acetic acid, 2,3,5-triiodobenzoic acid, 3,5-diiodo-4-amino-2-hydroxybenzoic acid).

Batelle Memorial Institute, Columbus, Ohio: Normal and iso-propanes, butanes and hexanes, S-alkyl thiouronium halides (S^{35}), pentaerythritol, and tetraethyl lead (C^{14}).

University of Illinois, Urbana, Ill.: Butadiene (C^{14}), aspartic acid (C^{14}), and 1,2 ring-labeled aniline, benzamide, phenylhydrazine, and tryptophan.

Tracerlab, Inc., 130 High St., Boston 10, Mass.: Ethylene oxide, 20-methyl cholanthrene (11-C^{14}), testosterone and cholestenone labeled in ring A, 2-labeled amino acids (leucine, norleucine, proline, tyrosine, valine, ornithine, etc.), vitamin A, dl-adrenaline, ethyl oxaloacetate (2-C^{14}), malic acid (2-C^{14}), and citric acid (1-C^{14}).

Southern Research Institute, Birmingham, Ala.: Pyrimidine and derivatives (pyrimidine nucleus labeled in the 4 and/or 6 positions), purines and derivatives from 2,4,6,8-C^{14}-labeled pyrimidine [adenine (2-C^{14}), guanine (4-, 6-, or 8-C^{14}), 2,6-diamino-purine (8-C^{14}), and hypoxanthine (2-C^{14})], and pteridine and its derivatives [folic acid (pteroyl-2-C^{14}-glutamic acid) diiodofolic acid (I^{131}), and diiodo-A-methopterin (I^{131})].

Radioactive Products Inc., Detroit, Mich.: Normal and iso-octane; and 1,6,11-labeled oleic and stearic acids and their derivatives (triglycerides, etc.)

Detroit Institute of Cancer Research, Detroit, Mich.: Incorporation of C^{14} into ring A of estrone and estradiol, and into rings A, B, C, and D of progestin and estriol.

Abbott Laboratories, N. Chicago, Ill.: Fluorescein and acridine type dyes, sulfanilic acid and As, Sb analogs (S^{35}), methionine (S^{35}), cystine (S^{35}), cysteine (S^{35}), barbiturates and thiobarbiturates (C^{14}), salicylic and gentisic acids (C^{14}), diiodotyrosine and thyroxine (I^{131}), penicillin derivatives (I^{131}, S^{35}), ascorbic acid (C^{14}), labeled blood constituents (I^{131}), organometallic derivatives (Co^{60}, Sb, Sr^{90}, As), pipsyl chloride (I^{131}, S^{35}), and yeast culture fractions (S^{35}).

United States Testing Company, Hoboken, N. J.: Glycerol, glycerophosphates, glycerides, atropine, *p*-amino salicylic acid, isopropyl alcohol, and acetone.

Temple University Research Institute, Philadelphia, Pa.: Oxygen-18 enriched labeled compounds (cuprous oxide, water, carbon monoxide and dioxide, methyl and ethyl alcohols, and formic and acetic acids).

National Bureau of Standards, Washington, D. C.: These chain-labeled sugars: glucose (2-C^{14}, 6-C^{14}); mannose (2-C^{14}, 1,6-C^{14}, 2,5-C^{14}); fructose (2-C^{14}, 2,5-C^{14}, 1,6-C^{14}, 1-C^{14}); idose (6-C^{14}).

THE SIXTH INTERNATIONAL CONGRESS OF RADIOLOGY . . .

. . . was held in London from July 23–29, 1950. Because of a lapse of thirteen years since the previous congress, an unusually large number of papers and symposia were scheduled, and a record number of interested delegates attended.

NUCLEONICS here brings its readers three papers and a report on one of the symposia that were included in the program. Several additional papers from the Congress are scheduled to appear in one of the next issues of NUCLEONICS.

Advances in Radiobiology between 1937 and 1950*

In this survey, the author foregoes comment on individual experiments in radiobiological research to note the development of a fertile tendency to seek a biochemical explanation for cytological radiolesions

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SINCE the last International Congress of Radiology, radiobiology, a science of applied physics, has considerably widened its field by exploiting the recent technical advances in physical research, chief among which are the production of neutrons and of artificial radioisotopes. Radioisotopes especially have opened up an immense new field to experimenters who use ionizing rays (the only ones to be dealt with in this paper); but biological studies with X-rays and natural radioactive elements have not diminished on this account. Thus, this limited space would hardly permit reference to all the papers published during the last thirteen years. I shall, therefore, without giving references, restrict myself to summarizing the achievements that have aroused the greatest interest during this period or yielded the most remarkable results.

Earlier work, especially that done between 1922 and 1937, had yielded results which seemed to be final:

The discontinuous, quantum, action of radiation on the cell.

The identity of the mechanism of action of the whole group of radiations characterized by their energetic capacity for producing ion pairs in living material through which they pass.

The attributing of the biological effect to this primary ionizing action.

The similarity of radiolesions to cytological "targets," the destruction or inactivation of which requires a definite number of "hits" by ionizing particles.

The purely quantitative differences (explicable in terms of the different ion densities along the particle tracks) between radiations as dissimilar as an electron and a proton.

The physico-chemical interpretation of the biological action of radiation, on the other hand, had not been much advanced.

* Paper presented at the Sixth International Congress of Radiology, London, July 26, 1950.

A direct effect of radiation, achieved by the removal of an electron from a "biological" molecule by the passage of a charged particle, was an unsatisfactory explanation of the phenomenon whereby a single impact on the complex macro-molecule constituting a gene produced a clear-cut mutation. The possibility was considered of an intramolecular energy-transfer to the site of a weak linkage, the breaking of which was the primary photochemical reaction initiating the mutation.

However, the results of experiments, in which soft X-rays of high intensity and radon in aqueous solution were used with the object of inactivating insensitive microorganisms, brought to light the toxic action of the hydrogen peroxide formed under these conditions. The idea of an *indirect* primary effect on the cell by decomposition products of the surrounding water caused by the radiation (hitherto considered particularly for interpreting the effects of irradiation on small *inorganic* molecules) was then extended to biological molecules.

Since it appeared that hydrogen peroxide (the concentration of which does not increase indefinitely as it is decomposed in its turn by radiations) is only biologically effective if present in considerable amounts, the curiosity of investigators was brought back once again to the old question of an *indirect* action; it has led them to analyze every reaction which could possibly follow the ionization of extra- or intra-cellular water.

The transfer of the radiation energy, the absorption of which has resulted in the activation of water molecules and of dissolved oxygen, could be effected either through an oxygen ion or through a hydrogen atom plus a free OH radical.

Role of activated oxygen. Besides the decomposition of water by radiations, activation of free molecular oxygen, which is normally present in solution, also occurs. Activated oxygen may be one of the most important producers of the primary lesion by acting on immediately neighboring molecules capable of acting as oxygen acceptors, either by direct fixation or by dehydrogenation.

Role of free radicals. The essential process in the activation of water by absorption of radiations may consist in the production of hydrogen atoms and hydroxyl radicals by means of one or other of various reactions which have been postulated. From the point of their formation, starting from the negative and positive ions formed along the track of the particles, these radicals, which possess high chemical activity, might be able to diffuse away and (before recombining) to reach solute molecules which would be oxidized by OH or reduced by H.

This interpretation has led to a new explanation of two phenomena: (**1**) The protection which, in a solution to which a second solute has been added, the second solute gives to the first. It is thus that the inactivation of a virus suspension can be effected by a much smaller dose in water than in broth, or in water to which gelatine, serum, or even much simpler substances have been added. These additional molecules act as "protectors" by capturing some of the free radicals, or, by accepting oxygen and thus turning on themselves the effect of the irradiation attributable to oxygen ions. (**2**) The reduced effectiveness of radiations on concentrated solutions as compared with diluted solutions (observed long ago in various biological experiments, especially those in which a comparison was made of the sensitivity of substances in the dry and in the hydrated states). The phenomenon may be explained on the grounds that, in less concentrated solutions, the ratio of the numbers of ions formed—precursors of

free radicals—to that of the dissolved molecules is higher.

Discussion. From the foregoing, it seems legitimate to conclude that different modes of indirect action are going on simultaneously during an irradiation; a direct action, which there is no reason to neglect, is also present. A fragile molecular structure cannot remain indifferent to loss or displacement of an electron belonging to the electron pair of an important linkage (a phenomenon likely to occur along the trajectory of an ionizing particle).

The remarkable progress made in the problem of the mechanism of the action of radiations *in vitro* has revealed the complexity of the phenomenon. It was studied, however, under the simplest experimental conditions, with known, isolated, and stable molecules.

Let us apply these different modes of action to the living cell. What is going on among these numerous, varied, and complex constituents, which constitute the cell and undergo in it perpetual transformation and re-formation, and about which little is known?

It is probable that all these mechanisms intervene and that, besides direct action, a principal primary effect may be attributable to activation of the cellular cytoplasm, a medium where water molecules largely predominate. Essential organic constituents are therefore probably reacted on by (*a*) oxyreduction, the free radicals having been formed at a distance of a few mμ; (*b*) molecules of activated oxygen, the probability of collision depending on the probability of their formation in the vicinity of a reactive molecule, the radiosensitivity of the latter being explicable by their quality of oxygen acceptors either by dehydrogenation or by oxygen fixation; and (*c*) even peroxides formed in the course of the irradiation.

We are unable to imagine the multitude of both the chemical reactions initially produced and those which arise from them during the course of metabolism of the affected molecule.

Repair of Certain Radiolesions

It has been found that certain radiolesions, in various cell species, are reparable spontaneously or artificially. The sequence of the reactions, which ordinarily leads to some definitive change or to death, may be interrupted or even reversed.

Lowering the temperature immediately after irradiation, by slowing down metabolic processes, gives time for the reparative processes to manifest themselves. The death rate of irradiated ova and yeast cells has been reduced in this way. Again, in the case of ultraviolet irradiation, spores of fungi and of various other microorganisms exposed to visible light show a considerable degree of restoration. The mechanism of this action is here even more difficult to interpret.

Alteration of Some Cellular Functions by Radiation

Much work has been done and important progress made in connection with cellular radiation-induced injuries that give rise to morphological or functional changes. The results have been interpreted in the light of new concepts in cytology by taking into account such chemical entities as the centers of enzyme formation in the cytoplasm and the cycle of nucleic acid exchange between nucleus and cytoplasm. This work illustrates one of the many fruitful applications in radiobiology of the tracer method with radioisotopes.

Cellular respiration. We must consider cellular respiration as one of the first functions to be disturbed, certain enzymes involved in respiration being rapidly inactivated by irradiation, especially those containing —SH groups. But blocking of these by oxidation would be partially reversible under the

influence of glutathione or of catalase.

Reduction of basal metabolism by irradiation has been demonstrated in rats by the use of C^{14}. After exposing whole animals to X-radiation, an increase is observed in the uptake of C^{14} in the organs coincident with a diminution in the $C^{14}O_2$ exhaled. But, while the C^{14} is retained in the tissues in the protein molecules, fixation of the element in the purines, particularly in those of desoxyribonucleic acids, is greatly reduced.

Cellular division. What has just been said confirms the observations made by means of ultraviolet microphotography, controlled by histochemical techniques. In radiosensitive tissues, one can follow, by these means, the accumulation of ribonucleotides in the cytoplasm of the cells exposed to X-rays, coincident with an inhibition of the synthesis of desoxyribonucleic acids in the nucleus. The arrest of mitosis, one of the first definite manifestations in an irradiated tissue, could then be explained by the inactivation of the enzyme which catalyzes the synthesis of new molecules of pentose nucleoproteins.

Work with P^{32} injected into rats with grafted sarcomas has shown a lower radioactivity in nucleic acids extracted from tumors previously exposed to X-rays. This decrease which affects more particularly the thymonucleic acid fraction can be attributed to a slowing down of its metabolism rather than to any change in permeability of the cell membrane to phosphate ions, which is not diminished to any extent by radiation.

The fact that the synthesis of the principal constituent of the nucleus can easily be disturbed by radiations helps us to understand not only the action of radiation in blocking cell division but also the various anomalies, visible or latent, of succeeding divisions, including the mutations which derive from it.

Discussion. It is interesting to compare these effects of ionizing radiations on the cell with the analogous effects brought about by chemical substances, such as urethane, an anaesthetic which also causes a diminution in basal metabolism (demonstrable by the method of C^{14} fixation). It has been established further (*a*) that with such generally toxic substances as urethane, mustard gas, and some of its derivatives, chromosome breaks, gene mutations, and even the induction of malignant processes can be obtained, and (*b*) that a protection effect against X-rays is particularly clear with substances containing certain active groups, especially —SH groups, while dithiol compounds protect against the attack of toxic vesicants on the free —SH groups of proteins.

All these facts, as well as others, seem to justify the application of the description "radiomimetic" to a series of chemical compounds tested in the treatment of cancer, simultaneously or conjointly with radiotherapy.

It is well, however, not to be unduly misled by this suggestive comparison. Even if it confirms the fact (known for a long time) that the apparent effects on cells and the morphological changes which they undergo during post-mortem disintegration, are, in general, the same for each species and vary little whatever the injurious agent may be, penetrating radiations nevertheless represent a unique mode of attack on the cell. The initial phenomenon, a purely physical one, has the exclusive property of affecting cells equally and of being able to bring into play every molecule of every compound, since it depends solely on the probability of absorption of radiating energy; the chemical and biological manifestations are only secondary.

With chemical agents, on the other hand, the initial phenomenon is a biological one. The permeability of the cell membrane to the toxin varies with

the cellular species acted upon and with the particular substance used.

In their turn, radiations, in the same way as the other varied modes of attack on the organism, elicit secondary nonspecific adaptation reactions. We should be careful in this connection to avoid any confusion which the expression "radiomimetic" might produce.

Effects of Total Radiation

Where the various mechanisms previously considered on the cellular level act simultaneously on all the elements of an organism, the complexity of the reactions may defy analysis.

This old subject, revived because of the high intensities of the penetrating rays (gamma rays and neutrons) which may be encountered, is being studied in a large number of radiological laboratories. Has not medical research the inestimable privilege of approaching without misgiving any subject, however crude in its origin, since its mission is always to counteract the misdeeds of nature or of man?

The most striking of the syndromes brought about in mammals by total body radiation is that described in March, 1922, as the "roentgen purple syndrome." Recognized as one of the principal causes of death in individuals exposed to general irradiation, the study of its mechanism, of its prevention, and of its treatment has been the subject of many recent publications.

To what, in the first instance, shall we attribute the most conspicuous symptom to attract special attention in recent years, i.e., the multiple petechiae which sometimes amount to a true hemorrhage? Shall we attribute it to the diminished coagulability of the blood or to the increased fragility of blood vessels? Various substances figure as factors in the delicate physiochemical reactions that occur to bring about the transformation of fibrinogen into fibrin: enzymes, vitamins, salts. Of diverse origin, and all essential for the success of the final result, they arise in such differing organs as liver, bone marrow, the intestine, and the reticuloendothelial system.

For a long time there has been a tendency to attribute the difficulties of coagulation and of the hemorrhages to precipitation of globulin following the destruction of megakaryocytes. Recent research has disproved this sequence by demonstrating that the peak of thrombocytopenia does not coincide with the hemorrhagic symptoms. On the other hand, the prothrombin level remains unchanged; and there appears to be no deficiency in thromboplastin, calcium salts, phosphates or fibrinogen.

Alternatively, the presence in the blood of an excess of "heparinoid" substances has been demonstrated in irradiated dogs. Prolongation of the survival time of such animals by the administration of "antiheparin" substances (notably toluidine blue) would seem to corroborate the evidence for the discharge as a result of irradiation of anticoagulant substances.

As soon as the production of histamine by the action of radiation was recognized, an appropriate role was assigned to it in the syndromes against which the effects of various antihistaminic substances have been tested. Among these must be reckoned desoxycorticosterone. But their administration has not as yet increased the survival rate.

Further, substances capable of affecting the fragility of blood vessels have been tried simultaneously. The best results would appear to have been obtained with flavin derivatives, especially with rutin, which has cured the hemorrhagic syndrome in dogs. However, this initial and encouraging result has not been obtained with other animals. Moreover, substances that control blood pressure (such as adrenalin

and adrenochrome) and those that are capable of counteracting vascular fragility (especially ascorbic acid) have been proved ineffectual, even when administered immediately after total body irradiation.

On the other hand, it has been possible to obtain preventive action with several substances, which were chosen because it was expected that they would have an antioxygen protective effect on enzyme systems. These results support the mechanisms, considered in the preceding paragraphs, which have stressed production of ionized oxygen and of peroxides as important agents for producing the initial radiolesions. Cyanides, tocopherol, and cystine, administered a measured time before irradiation, have produced a considerable decrease in the mortality rate by directly preventing the oxidation of molecules essential for metabolism, or by inhibiting peroxide formation.

Conclusions

In this survey, it has been necessary to omit highly interesting experiments on the application of radiobiology to the solution of particular problems, owing to their great number and diversity. An attempt has been made rather to show the development of a tendency, apparently a fertile one, to seek to provide a biochemical explanation of cytological radiolesions. The use in such investigations of more powerful tools such as the electron microscope and other apparatus yet to come will render this synthesis of the research on the molecular level a still more intimate and profitable one.

*Report of "Symposium on Radiation Chemistry"**

The inclusion of a radiation chemistry symposium in the program of a Radiology Congress is indicative of the great extent to which essentially chemical ideas have pervaded fundamental radiobiology

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The Symposium on Radiation Chemistry was intended mainly as a review of recent developments for the benefit of biologists and radiologists who were not familiar with the field. The topics covered in the symposium can be divided into (*A*) an historical introduction to the mechanism of "indirect action" of ionizing radiations in solutions; (*B*) a review of recent experimental work bearing on the mechanism of indirect action; (*C*) a discussion of the possible mechanism; and (*D*) a general discussion (on units).

Mechanism of "Indirect Action"

In opening the symposium, *Dr. C. B. Allsopp* remarked that the action of ionizing radiation on living organisms has always suggested that a chemical process was involved at some stage in the production of the effect, even when the results could be explained by the "target theory." In this sense, a

* Held at the Sixth International Congress of Radiology, London, July 25, 1950.

general theory of radiation effects will always be to some extent a chemical theory.

One such theory has emerged from the study of the action of radiation on substances in dilute aqueous solution. This is the theory of indirect action (*1*). It may be said to rest on three fundamental developments since 1937.

1) The discovery of the "dilution effect" (*2*), in which the yield of indirect action is independent of the concentration.
2) The discovery of the "protection effect" (*3*), in which the yield may be lowered by the addition of a second substance.
3) The suggestion that the active chemical constituents of irradiated water might be H atoms and HO radicals (*4*).

These ideas suggest a mechanism by which the energy absorbed in irradiated solutions can diffuse through the solution in the form of H atoms and HO radicals.

Recent Experimental Work

Dr. W. M. Dale said that though his original experiments on the protection effect (*3*) could be explained as a simple competition between two solutes for the active constituents of irradiated water, his more recent experiments (*5*) had revealed that the phenomenon was more complicated than originally supposed. This was demonstrated by a study of the specific protective power of a number of substances.

The degree of protection afforded by x grams of "B" to y grams of "A" when both were in solution had been found to depend upon the chemical constitution of "B." It was later found that the specific protective power of "B" often decreased continuously with increasing concentration of "B" over the entire range of concentrations studied. This implies at least one other process besides competition.

Dr. Dale also noted that some compounds do not show the dilution effect in that there is no range of concentrations in which the yield of indirect action is independent of the concentration. The yield of deamination of glycine by X-rays increases with the concentration of glycine up to saturation (*6*). The same phenomenon is observed in the liberation of sulfur from thiourea.

Dr. Walter Minder described some experiments in which the acid HX was formed from organic halides of the type RX when the latter were irradiated in dilute aqueous solution (*7, 8*). The yield of acid was found to depend upon the structure of R, being invariably lower when R was aromatic and higher when R was aliphatic. The effect is characteristically indirect in that the yield of HX is independent of the concentration of RX over a wide range. From the lower limits of this range, it can be deduced that the maximum diffusion distance of the active constituents of irradiated water varies from 20 to 100 water molecule diameters, depending upon the substance RX studied.

The same effects were observed in alcohol (*9*) and acetone solutions. From this fact it can be inferred that an indirect effect is operative in three solvents as well.

Dr. P. Bonet-Maury discussed the production of H_2O_2 in water irradiated with X-rays (0.9 A.U.) and alpha particles (*10, 11*). The analytical technique was sensitive to 0.1 γ per ml. In the case of X-rays, the yield of H_2O_2 is a complicated function of the pH and increases with the oxygen concentration. No H_2O_2 could be detected in water freed from oxygen. The production of H_2O_2 by alpha particles is insensitive to the pH and completely independent of the oxygen concentration. The kinetics of H_2O_2 production also vary characteristically with the intensity for each type of radiation.

These results can be attributed to a difference in the initial distribution of H atoms and HO radicals produced in water by the two types of radiation. In the case of X-rays, the initial distribution seems to favor an unproductive recombination of the radicals. This process is inhibited by oxygen, possibly by the reaction

$$H + O_2 \rightarrow HO_2$$

with subsequent formation of H_2O_2 by

$$HO_2 + H \rightarrow H_2O_2$$
$$HO + HO \rightarrow H_2O_2$$

In the case of alpha particles, the initial distribution favors the production of H_2O_2 by the last reaction above and is uninfluenced by the oxygen concentration, at least up to the maximum concentrations studied.

The Possible Mechanism

Dr. F. S. Dainton reviewed the experimental evidence concerning the relative abundance of the ions H_2O^+, HO^+, H^+, and H_2O^+ formed in water vapor by electron impact. He also discussed the average energy associated with the production of each species. He suggested that, in liquid water, about 16 ev might be used for ionization and 14 ev used for excitation of water molecules.

The most abundant ionic species are probably H_2O^+, formed by primary ionization, and H_2O^-, formed by subsequent electron capture. The excited water molecules dissociate into H atoms and HO radicals, but in the liquid phase the yield of atoms and radicals is diminished by recombination. However, any dissociative process which can be facilitated by the solvation of ions or radicals will be favored, e.g.

$$H_2O^+ + H_2O \rightarrow H_3O^+ + HO$$
$$H_2O^- \rightarrow HO^- + H$$

The presence of the ions has not been demonstrated, but the presence of free radicals can be shown by adding to the water a small amount of an organic monomer which can be polymerized by the radicals (*12*, *13*). Irradiation of such a solution produces an adundant precipitate of the polymer. It can be shown that the polymerization is initiated by HO radicals.

A study of the kinetics of polymerization by X- and gamma rays compared with kinetics induced by HO radicals produced in the photolysis of H_2O_2 suggests that, even in the case of these sparsely ionizing radiations, the initial distribution of the radicals is nonuniform.

It can be concluded that the radicals must initially be concentrated along the tracks of secondary electrons projected in the solution by Compton and photorecoil processes.

Dr. Milton Burton said that, in considering the effect of ionizing radiation on a living organism in aqueous suspension, several possible mechanisms should be recognized. The track of the ionizing particle may pass through the organism or may pass close enough to it to produce an indirect effect. In the first case, the organism will be ionized as follows: $M \rightarrow M^+ + e$. Eventually M^+ will be neutralized by a slow electron, giving rise to an electronically excited state, M^*. M^* may decompose spontaneously, may react with the circumambient layer of water, or may be deactivated without chemical change.

If the track of the ionizing particle passes through the water near the organism, the organism may be affected by ions and oxidizing entities produced in the water, H_3O^+, HO^-, HO, HO_2, H_2O_2 (see above). Any of these substances in local concentrations near the organism may result in damage and possible eventual death. Of these, HO_2 is probably the longest lived.

The "target area" of the organism is, therefore, determined not only by the geometrical cross section of the organism to ionizing radiation but also by the area of the circumambient water in which the organism can be damaged indirectly. The target hypothesis is

also complicated by the possibility that a hit sustained within the organism may not prove fatal.

Dr. Burton indicated that these considerations might lead to the observed relation between the measured target size and the target size calculated from the results of irradiation (*1*).

Discussion on Units

In the general discussion which followed, it was agreed that an unfortunate feature of the experimental work in radiation chemistry being published was the confusing multiplicity of units used to express radiation chemical yield.

The most desirable unit is one expressing the amount of chemical change per unit amount of energy absorbed in the reacting system. The simplest unit in common use is G, the number of molecules changed per 100 ev of radiant energy absorbed in the reacting system. The choice of 100 ev as the unit energy usually makes G greater than 1. It was pointed out that the total energy absorbed could usually be measured with fair precision, whereas a calculation of the number of ion pairs formed involved an assumption that was often invalid.

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*Protection Against Radiation Hazards**

Here summarized, in a general review, is the status of our present knowledge of the effects of ionizing radiations on humans and animals. This is recounted in relation to the necessity for certain precautions

By SHIELDS WARREN† and AUSTIN M. BRUES‡

In protecting the body against ionizing radiation, we are dealing with a complex of cells, the components of which vary in their threshold of response.

Four variables in the organism must be weighed: 1) the susceptibility of the tissue; 2) the proliferative activity of its component cells; 3) the metabolic activity of its component cells; and 4) the mass of tissue irradiated.

So far as the radiation is concerned, we must consider: 1) the character of the ionizing path; 2) the amount of radiation; 3) the rate of the delivery;

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and 4) the duration of periods of respite from radiation.

Varying tissue susceptibilities are dramatically shown in the nature of the acute total body radiation response itself, where it would appear that the major factors leading to morbid effects are the changes in the blood-forming organs and the intestinal epithelium. The radiation therapist is familiar with other tissue peculiarities which show up as a result of localized irradiation.

Proliferative activity of cells is ordinarily associated with radiosensitivity, perhaps because of the special sensitivity of the dividing cell to destruction. But, in some cases, regeneration after injury may be conducive to rapid recovery, as in the erythropoietic system after phenylhydrazine (*1*) or the myelopoietic system a few days after estrogen treatment (*2*).

Metabolic activity, in addition to its effect on isotope deposition, is important in the pure radiation effect, for example, in the prophylactic value of anoxia or cysteine treatment during irradiation. Low temperature, on the other hand, merely delays the onset of the lethal picture.

Dose-rate effects are of great interest but cannot be discussed here. By analysis of dose-rate relationships for radiation of low and high ionization densities (e.g., X-rays and neutrons), it may be possible to analyze many mammalian responses into their basic components.

The Protection Problem

In light of these factors, the problem of protection can be divided into several phases:

1) Protection against acute radiation from external sources
2) Protection against repeated or continuing irradiation from external sources
3) Protection against acute radiation from internal sources
4) Protection against acute or repeated radiation from internal sources
5) Protection against any combination of the above.

In general, radiologists are primarily concerned with protection against repeated or continuing radiation from external sources. As therapeutic appliances tend toward more penetrating roentgen radiation in therapy and to some degree even in radiography, the danger of exposure of the hematopoietic tissue becomes greater than that of exposure of the skin owing to the greater sensitivity of the former.

Maximum Permissible Dose

The permissible dose in use in installations of the U. S. Atomic Energy Commission and in general in North America is 0.3 roentgen per week calculated at the depth of (5 cm beneath the skin) the major hematopoietic tissues. Conversely, since the mass of hands and arms is small relative to the whole body and since no hematopoietic tissue is present in them, their permissible dose is 1 r at the basal layer of the skin.

For protection purposes it must be assumed that the total body is irradiated, although partial shielding by sinks, metal furniture or other objects may actually materially reduce the hazard. Some radiologists even now are insufficiently alert to the danger associated with use of their apparatus and depend too much on infrequent blood counts for warning of danger.

Continuous area monitoring or repeated checks of equipment and rooms is the only sure way of being safe. The use of film badges or blood counts serves to show only what has happened and, while useful as a check, serves only as that. A pencil ionization chamber examined periodically has more value.

In area monitoring, it is important to remember that most monitoring instruments are planned to detect accu-

rately rather specific ranges of radiation and, unless planned and calibrated for the particular type of radiation, may give misleading data.

Effects of Overexposure

In guarding against external whole body irradiation, it is important to keep in mind what we are trying to prevent happening. First and most common is radiation dermatitis, often accompanied by ulceration and neoplasia. Second, is damage to hematopoietic tissue, manifest as either agranulocytosis, anemia, or, more rarely, leukemia. Intensities sufficient to produce widespread radiation osteitis would in all probability be fatal before this lesion had time to develop.

So far as the gonads are concerned, we must still rely on speculation in part. Sterilization by total body radiation seldom occurs, because the dose required is usually as much or more than the fatal dose of total body radiation in the male, and the dose required for sterilization in the female is even greater. As has been pointed out by Muller (*2*), gene mutation resulting from exposure of gonadal tissue to ionizing radiation is a very definite possibility. So far as our knowledge goes, no gene mutation has been induced by radiation that does not also occur spontaneously. As Muller indicates, however, the rate of either spontaneous gene mutations or irradiation-induced mutations are so low per locus that no really effective comparisons as to rate can yet be made.

There is a considerable degree of proportionality of gene mutation rate to the dose of X-rays, and from the evidence in hand it appears that even the smallest doses of radiation may have their share of mutagenic effectiveness. It is quite possible indeed that even a single ionization may be sufficient to cause a mutation, since no evidence has been found for the existence of a threshold for mutation production.

In dealing with protection, not only must gene mutations be weighed but, in addition, the possibility of chromosomal aberrations must be considered. There is some evidence that, in mammals, most chromosomal aberrations result in sterility or semisterility. Semisterility, in particular, in view of the breeding habits of man, is not too significant an event.

The practical experience of radiologists over the years warrants the conclusion that the present standards of radiation protection are adequate when due regard is paid to safety regulations. It is to be hoped that further studies in human and mammalian genetics will enable us to clarify these points even further. As yet, frequency of gene mutations induced by a given dose of radiation has not been adequately determined. It is hoped that experiments currently underway on mice at Oak Ridge and elsewhere will throw additional light on this point.

Internal Radiation

In contrast to external radiation which falls upon the "just and unjust alike," internal sources of radiation may be highly specific in their localization. With the large number of radioisotopes and the varying body responses of the metabolic tissue to different elements, it is possible to obtain an almost infinite number of variables. Consequently, the problem of controlling internal radiation is far more complex than that of external radiation.

The metabolism of the isotope, its emission spectra, its half-life, its biologic half-life, and its localization or concentration are all of great importance. So far as we can judge in the case of internal emitters, the sensitive tissues, the indicators of reaction, are the hematopoietic tissues and the bone, in general, although there are other instances where specific uptake introduces additional factors as, for example,

the specific localization of iodine in the thyroid.

Isotope Hazards

Examples of a few radioelements will serve to illustrate the range of factors involved in the calculation or estimation of isotope hazards. In the case of sodium-24, the physical half-life is the important factor. Because of the homogeneous radiation flux to which the body is subjected after ingestion of Na^{24}, the effects are qualitatively similar to those of total body irradiation. As a result of physical decay (the half-life is 14.6 hours), the radiation dose is distributed in tissue accordingly, since the rate of loss by excretion is relatively very slow. The lethal dose (calculated in physical equivalents) is slightly greater than that of a single dose of X-ray, largely due to its distribution over a few days.

With sodium-22, on the other hand, the total body dosage depends primarily on the turnover rate of sodium in the body due to excretion, since the half-life of this isotope is 3 years.

Phosphorus-32 is a radioisotope whose general clinical effects are a little different from those of total body irradiation. Its physical half-life (2 weeks) largely determines the duration of its effectiveness, the loss of radioactivity being somewhat accelerated by metabolic loss. The geometrical pattern of irradiation from this beta-emitter is determined by its deposition by metabolic turnover, in tissues high in phosphorus, and especially in tissues that are in an active state of growth. Such tissues in turn, by synthesis, deposit P^{32} even in those compounds (such as desoxyribose nucleic acid) that are rather metabolically inert in nongrowing tissue.

The isotope also exchanges readily with the phosphorus in bone phosphate. The result is somewhat selective irradiation of bone marrow and blood-forming organs. Certain blood dyscrasias, including polycythemia rubra vera and some of the leukemias, are effectively treated by P^{32}, but it is suitable only for such conditions as are radiosensitive, since it is selective also for the normal blood-forming organs which play a major role in the etiology of radiation sickness.

Friedell (*4*) has shown that bone tumors can rather consistently be produced in rats given a sufficiently high dosage of P^{32}, but they have not, so far, been observed in treated humans.

The isotopes that are primarily bone-seekers require special attention, since a large number of elements, including many of the fission products, fall in this class. In other cases (e.g., C^{14} carbonate), the critical dosage may well be delivered to the bone. The alkaline earths (calcium, strontium, barium and radium) are best understood, and indeed our only significant clinical experience is due to the acquisition of radium, with or without mesothorium. Other radioisotopes which concentrate in bone include the rare earths, plutonium and other transuranic elements, and gallium. Because of the susceptibility of bone to carcinogenesis, none of these elements has shown promise in the therapy of bone tumors, although concentration in actively growing bone tumors may somewhat exceed that in normal bone.

The pattern of bone tumor development (*5*) has been worked out in the case of sustained administration of strontium-89. Within the life-span of the mouse (experimental material which can be used in large quantities), it appears that a given dosage rate results in a continuously increasing morbidity rate throughout life. Whether this increased rate would continue indefinitely is not clear and Evans* has suggested that the morbidity rates observed are actually the early phases of Gaussian probability curves, such as

* See footnote on next page.

one sees with random distribution of events around a mean-time interval. Such a random picture is actually seen in the case of X-ray-induced mouse lymphoma, with the maximum incidence about 200 days after treatment, the effect then subsiding.

The answer to this question can have profound influence on our estimation of the hazards of a large number of isotopes, but it will have to be derived from experimental work on longer-lived animals. On the other hand, it may become available from study of data on human radium poisoning, since it is quite possible that the peak incidence of radium sarcoma may have passed with the third decade following the ill-controlled early exposures.

Another question about which we have essentially no knowledge is whether there is a threshold amount of bone radioactivity below which bone tumors will not be induced. It is a fact that both human and animal bone sarcoma is not usually seen in the absence of bone necrosis, yet the evidence does not rule out the possibility that cancer, like the genetic mutation, may be a random event with a small probability of resulting from a single ionization.

It may still be true that cancer results from a cellular change occurring in a partially devitalized tissue. This question may be settled as a result of further experimental work, or possibly from observation of populations receiving diverse background radiation attributable to differences in environmental radium abundance.

The results of bone irradiation may be quite different, depending upon the localization of the offending element. In the case of plutonium-239, it has been estimated that this element is (per ionization) about fifty times as toxic to small animals, both acutely and chronically, as radium (*5*). Since the toxicity of plutonium is entirely due to the dense ionization of alpha particles, and radium is largely due to it, the difference is apparently mainly due to localization of the radiation (and partly to the higher retention of Pu^{239}). As to acute toxicity, the difference may be due to the early concentration of Pu^{239} in blood-forming organs. The late toxicity, being largely a matter of bone pathology, is quite likely attributable to the highly selective concentration of Pu^{239} in the osteoid matrix in the endosteal and periosteal regions from which, no doubt, the most metabolically active bone cells are irradiated. Radium, the calcium *analog*, on the other hand, deposits to a considerable extent in the more metabolically inert areas.

Human permissible levels of the bone-seeking elements are necessarily adjusted on the basis of the human radium experience. There are a few data on bone tumors resulting from X-irradiation which indicate that 1,500 r or more may be carcinogenic to bone, but it is desirable (since it is practically impossible to calculate bone radiation dosage from a heterogeneously distributed element) to use comparative data with radium in animal experiments.

The present inclination is to consider that one microgram of radium deposited in the human skeleton is potentially dangerous; but re-examination of the human toxicity data has been discussed by Dr. Evans.* This is very important in view of the considerable number of isotope levels which are pegged to the human radium experience.

No discussion of isotope toxicity would be complete without some consideration of the possible influence of the finer distributions of radioactive particles on the genesis of cancer. Whether a small total amount of radia-

* Discussed by Robley Evans in paper entitled "Quantitative Aspects of Chronic Radium and Mesothorium Poisoning and Their Relation to the Establishment of Maximum Permissible Doses," presented earlier at the symposium.

tion is more likely to be carcinogenic if it is concentrated in a few areas of a susceptible tissue is a question that remains unsettled.

A particularly interesting isotope, from the standpoint of toxicity, is carbon-14. Our knowledge of this subject is limited to carbon dioxide and the most labile carbon compounds, but since attempts at radiation therapy by carbon compounds must be preceded by an understanding of toxicity, this work is of great importance.

It has been established that C^{14} in ambient CO_2 exchanges very efficiently with bicarbonate of blood and tissues and with part of the bone carbonate, even though the gross metabolic flow of CO_2 is in the opposite direction (*6*). The carbon in soft tissue, which makes up the bulk of the body carbon, on the other hand, presents a much different picture. Buchanan (*7*) has shown that a rapid exchange between $C^{14}O_2$ in air and blood bicarbonate takes place during the first week of continuous exposure, but that the protein and other carbon in soft tissue then equilibrates at $\frac{1}{50}$ or less of the specific activity of the alveolar air CO_2 or blood bicarbonate. This indicates that the remainder of the tissue carbon is formed from foodstuffs.

A quite different picture must occur when C^{14} is ingested in a labeled diet. As in the case of P^{32}, carbon taken into growing cells becomes fixed more firmly, and is very slowly lost. In the case of injected labile C^{14} compounds (as well as bicarbonate), the bulk of the C^{14} is lost in the expired air (up to 98 or 99 % in the first day), but even here the relatively slow metabolic rate of the human being, which is about $\frac{1}{10}$ that of the small rodent, indicates that further experimental work will be necessary before a good picture of human C^{14} fixation can be established.

In looking forward to the optimal use of radiotherapeutic agents, we shall have occasion to collect much information on the various characteristics of isotopes which have been mentioned, and shall, in addition, welcome any knowledge of the treatment of acute or chronic radiation sickness.

Radioactive Wastes

The sources of exposure to internal radiation are now, fortunately, relatively few. The dangers of the promiscuous use of radioactive water have been recognized, and it rarely appears on the market. Accidental ingestion in the laboratory, therapeutic application, and contact with radioactive wastes represent the great majority of problems.

From the standpoint of radioactive wastes, which may be produced in considerable amounts by chain-reacting piles, hazards, with the exception of those created by the presence of the isotopes in air or in drinking water, can be relatively well controlled. In regard to radioactive wastes, one has to accept a dual standard: one for those persons in the laboratory or plant who are working under controlled conditions and in whom the extent of uptake of material can be accurately determined to prevent the dangerous level being attained, and another for the general population, where the need for detailed protection and careful study of the individual is not indicated.

In light of this, the aim of those supervising waste disposal should be to maintain the concentration of the wastes at the lowest possible level.

Indeed, some of the standards actually seriously proposed are lower than those occurring naturally in some of the rivers that are used for the water supply to heavily populated areas in the United States. It is essential that a sense of realism and balance be maintained and that we do not attempt to establish standards incapable of enforcement. As data accumulate on the

naturally occurring sources of radiation in drinking water, much more intelligent levels can be set.

At present, a degree of caution is being exerted, and properly so, that is far greater than is used in measuring out doses of such useful, but deadly, drugs as digitalis or scopolamine.

As an example of the satisfactory way in which these extraordinarily low levels of radioactivity can be attained even in streams receiving large amounts of radioactive wastes, a considerable share of the drinking water for the population of Richland, Washington, is taken from the Columbia River only a few miles down stream from the effluent from the Hanford Works.

Owing to the great dilution effect in air, the possibility of radioactive materials' reaching even remotely diffuse levels at or near atomic-energy plants is virtually nonexistent. The development of atomic energy so far has shown that the hazard may be forseen, predicted and avoided by adequate study, research and attention to detail.

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The Betatron in Cancer Therapy*

Flat radiation distribution and deeper concentration of dosage in cancer therapy is possible with betatron-produced X-rays. Equipment used and physical data obtained with a 22-Mev betatron are discussed

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FOR MANY YEARS there has been a trend toward the use of ever higher energy X-ray machines in cancer therapy, because with higher energies it is possible to deliver more radiation to the tumor with less damage to the intervening layers of healthy tissue. In fact, many new types of X-ray machines designed primarily for nuclear physics have been utilized in the study and treatment of malignant disease.

Therapists for some time were limited to voltages of the order of 200 kv which could be produced conveniently by simple transformers in shock-proof installations. Voltage-doubling circuits have been used in a few cases to produce energies up to 1 million volts. Within the last 10 years the resonant transformer, the Van de Graaff generator and the betatron have enabled one to use even higher energies for therapy. The first

* Based on a paper presented at the Sixth International Congress of Radiology, London, July 23–29, 1950.

† Also Physicist to the Saskatchewan Cancer Commission.

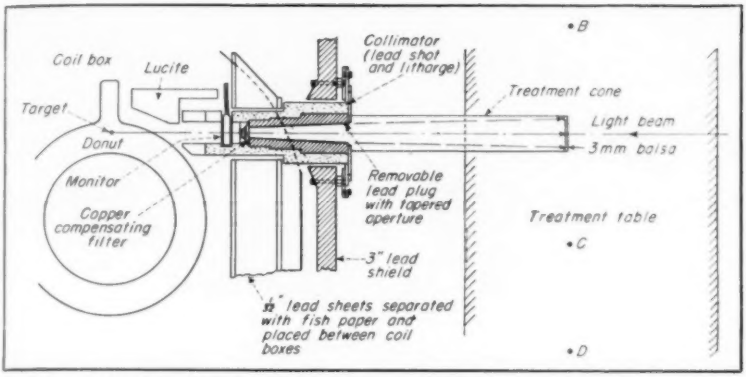


FIG. 1. Schematic diagram of the betatron showing the arrangement of monitor, copper compensating filter, collimator and shielding

two can be made to operate up to 2 million volts conveniently, and a number of such installations are in operation.

The betatron is also a device which has important clinical possibilities. This machine has proved very useful in the X-ray testing of heavy armament because of the great penetrating power of its beam. This same penetrating power makes it a useful device in the treatment of certain deep seated tumors.

Since April, 1949, a 22-Mev Allis-Chalmers betatron installed in the Physics Department of the University of Saskatchewan has been used to give over 50 patients full courses of X-ray therapy. The medical aspects of this work have been under the direction of Drs. T. A. Watson and C. C. Burkell of the Saskatchewan Cancer Commission. The clinical results for the first 36 patients are reported elsewhere (*1*). This betatron was the first used in the routine treatment of cancer patients, although the Illinois groups (*2*) used a similar machine about a year earlier to treat one patient.

Collimation of X-ray Beam

To make the high-energy X-ray beam from the betatron useful for medical work, it is necessary to be able to select any size radiation field conveniently, making sure that radiation outside this field is biologically insignificant. The intensity of the radiation over the area of the selected field should be approximately constant.

The method by which this is done is shown in Fig. 1. Collimation of the X-ray beam is attained by the use of a master collimator into which may be inserted any one of a number of lead plugs. The master collimator consists of a stainless-steel nonmagnetic rectangular case which is welded to a steel plate. It is filled with a mixture of lead pellets, lead oxide and glycerine, which sets quickly to give a smooth hard finish. The channel inside the collimator has two right-angle steps and is designed to take the lead plugs. It also holds in place the copper compensating filter, the monitor, and a light treatment cone which is fastened to the front plate of the master collimator. These components are shown in Fig. 2.

The master collimator is held in place by two bolts fastened to the 3-in. shield, which in turn is bolted firmly to the betatron. Some adjustment of the position of the collimator with respect to the betatron is possible. The axes

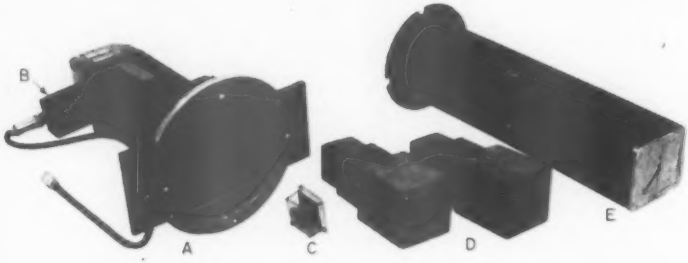


FIG. 2. *A*: Master collimator. *B*: Monitor inserted in the end of the collimator. *C*: Copper compensating filter. *D*: Lead plugs giving square and rectangular fields. *E*: Treatment cone consisting of a light-weight rectangular metal case fastened to a base plate and closed with a thin layer of balsa wood. This treatment cone may be fastened to the collimator with four wing nuts

of the master collimator, lead plug, copper filter, and treatment cone are made to coincide exactly. Adjustment of the position of the front plate of the collimator moves their common axis in space. The lead plugs can be made in a variety of internal sizes and may be inserted into the front face of the betatron. The betatron is designed so that a rectangular field 10 × 15 cm can be obtained at a distance of 105 cm.

Between the poles of the betatron are auxiliary shields of sheet lead separated by fish paper. Lucite blocks placed between the poles serve to reduce the intensity of scattered radiation by stopping electrons without the production of much radiation. This arrangement gives ample protection to the patient, the dosage rate outside the main field being about 0.05% of that within the main beam.

The light treatment cone serves merely to mark the point at which the field enters the patient and does not collimate the beam; the collimation is carried out by the lead plug. For each lead plug there is a corresponding treatment cone. The end of the treatment cone is covered by a 3 mm layer of balsa wood. The point at which the X-ray beam emerges from the patient is marked by a light beam which is directed from the far side of the room along the axis of the X-ray beam. The betatron arranged for therapy is shown in Fig. 3.

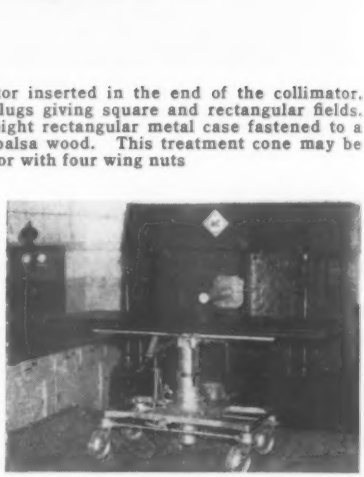


FIG. 3. Betatron in the Physics Department of the University of Saskatchewan, as arranged for cancer therapy

Copper Compensating Filter

X-rays are produced within the donut of the betatron when the high-energy

electron beam strikes the target. At these high energies the X-ray beam is highly concentrated in the direction which the electrons had just before entering the target. This concentration is so severe that the radiation reaching the edge of a 10 × 15 cm field at 105 cm is only about 50% of the intensity at the center of the field. A much greater treatment distance could be used to obtain a uniform field, but this would prove impractical because of the low dosage rate. A uniform field may be obtained by the use of a copper compensating filter shown in Fig. 4. This reduces the intensity along the axis of the beam and may be designed by trial and error to yield the flat distribution of radiation shown in Fig. 7.

A slightly different copper filter is shown in Fig. 2. A number of materials could be used for the compensating filter, but copper, with its relatively high density and low atomic number, is satisfactory. Carbon and aluminum, with lower atomic numbers, would be somewhat better but are rather unwieldy since they would have to be made thicker because of their lower density.

The spectral distribution of energy in the forward direction from the betatron, as determined theoretically by Schiff (*3*), is shown in Fig. 5, curve *A*. The radiation which emerges from the betatron is filtered by the donut and

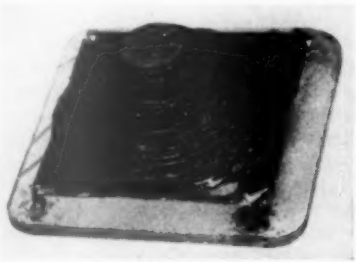


FIG. 4. Copper compensating filter

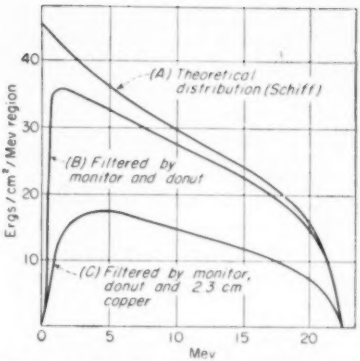


FIG. 5. Graph showing variation of energy flux with energy for a betatron operating with a maximum of 22.5 Mev

monitor and is given by curve *B* in Fig. 5. Curve *C* was obtained by computing the transmission at each energy by 2.3 cm copper, the thickness of the copper filter. At all energies above 4 Mev the fraction transmitted by this filter does not vary by more than 3% from an average value of 0.524.

It is seen that the copper filter does not alter the spectral distribution of the radiation; its absorption coefficient is almost constant over the energy range being considered. A carbon filter would tend to increase the portion transmitted at the high energies slightly. In the energy region around 20 Mev, carbon and other materials of very low atomic number show a slight decrease in the linear absorption with increase in energy. For these materials the increase in the pair production coefficient with energy is not quite as rapid as the decrease by the Compton process.

To achieve a flat distribution of radiation in practice, the direction of maximum intensity of the unfiltered beam must coincide with the axis of the filter. Relatively small errors in alignment can make the compensating filter useless. This is one of the difficulties in the use of betatron radiation for cancer therapy, as the alignment must be checked peri-

odically to insure correct operation. This adjustment becomes more difficult with very high energy betatron beams, because the collimation in the forward direction becomes enhanced with increase in energy. This narrowness of beam will probably prevent the use of energies of more than about 50 Mev for cancer therapy. It is doubtful if the slightly increased percentage depth dose of 50-Mev radiation will compensate for the greater difficulty caused by the "peaking" of the beam.

Calibration of the Betatron

The radiation intensity from the betatron is under continual observation at the control panel through use of the monitor shown in Fig. 2 and the basic electronic circuit shown in Fig. 6. An intensity indicator is necessary so that the betatron operator can continuously adjust injection and contraction timing to give maximum yield.

The monitor is an ionization chamber consisting of a series of parallel aluminum plates placed 2 mm apart and connected as indicated in Fig. 6. Current from the ionization chamber charges the capacitor C and flows through the high resistance R_1. The voltage drop across R_1 is applied to one grid of a twin triode used as a cathode follower. The radiation intensity is recorded on a 0–100 microammeter placed between the cathodes of the twin triodes. When the capacitor C charges to a predetermined value of about 30 volts, T starts to conduct. This fires a thyratron which actuates a counter, and also closes a relay which discharges C, allowing the process to start again.

To calibrate the machine, a Victoreen 25-r chamber is placed at the center of a 10-cm Lucite cube at the end of the treatment cone. The number of roentgens delivered for each discharge of the capacitor C can thus be determined. A value of about 3 r per count is satisfactory. The monitor integrates the same radiation regardless of the size of the lead plug which is inserted in the collimator, so the same calibration can be used for all of the treatment cones.

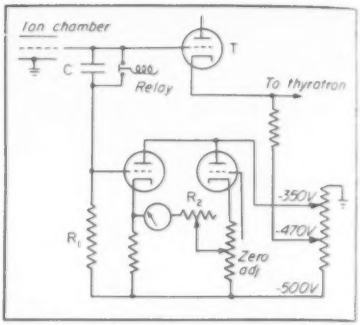


FIG. 6. Schematic diagram of the integrator used for the delivery of a predetermined dose with the betatron

Changes in the energy of the betatron produce slight changes in the calibration, for as the energy is increased, the radiation is collimated more in the forward direction and a slightly larger reading is obtained on the Victoreen chamber per count from the monitor.

The Victoreen chamber is placed at the center of the Lucite cube to build up electronic equilibrium between the primary radiation and the secondary electrons. Doses measured in this way have been called "roentgens," but it is doubtful if radiations of these energies can really be measured in terms of such a unit.

The dosage rate at the end of the treatment cone with the filter in place is about 70 r/min, whereas without the filter the dosage rate is about twice as great. The monitor, being much closer to the target, sees a dosage rate of about 1,600 r/min. This average dosage rate is produced by bursts of radiation lasting about 1 μsec, repeated 180 times per second.

The dosage rate within each burst is exceedingly high, and problems involving the recombination of ions become

important. From the capacity of C (Fig. 6) and the voltage required to actuate the tube T, it is possible to determine the number of ion pairs produced in the monitor per cm³ per burst. For an output of 150 r/min at 1 meter, this is approximately 3×10^8 ion pairs/cm³.

After the burst occurs, positive ions move in one direction, negative in the opposite. The region in which recombination can occur for each burst is thus a continually decreasing volume near the center of the monitor chamber. It can be shown that the percentage P of the ions collected is given by

$$P = 100x \ln \left(1 + \frac{1}{x}\right)$$

$$\text{where } x = \frac{v}{a^2 n_0} \frac{(u_+ + u_-)}{\alpha}$$

where v is the potential between the plates of the ionization chamber separated a distance a; n_0 is the number of ion pairs/cm³ per burst; u_+ and u_- are the mobilities of the positive and negative ions respectively; and α is the coefficient of recombination.

Substituting values for these quantities, it will be seen that 320 volts will collect 99% of the ions when the plate separation is 2 mm. Examination of the equation will show that if a is increased to 6 mm, nine times the voltage is required to collect the same percentage of ions. Before this voltage is reached, the ionization current increases rapidly because of secondary emission produced by ionic bombardment of the plates.

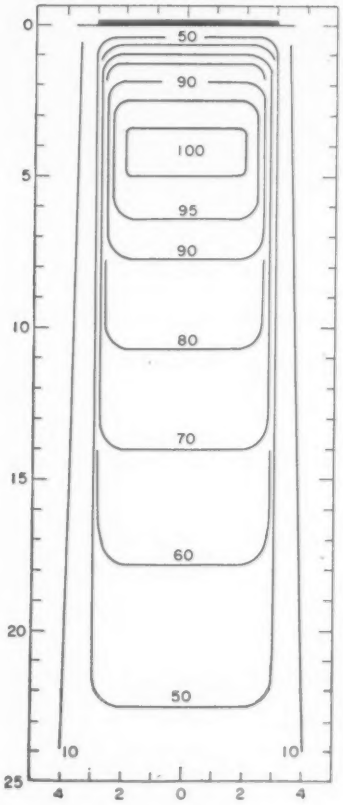
To be successful, therefore, the monitor must use plates which are separated by a small distance. A monitor using a series of parallel aluminum plates 2 mm apart has proved satisfactory. Since considerable variation in temperature occurs at the position of the monitor (Fig. 1), the monitor should be vacuum tight. Then its calibration remains independent of temperature and pressure. Using this monitor it is possible to deliver doses with an accuracy of 1 to 2%, which is clinically satisfactory.

Distributions of Radiation

In Fig. 7 is shown the distribution of radiation obtainable with a 6-cm diameter circular field using 22-Mev radiation at a focal skin distance of 105 cm with a copper compensating filter. The maximum dose obtained is received at a depth of about 4 cm; this dose has been called 100%. The doses at other depths are expressed as a percentage of this dose. It is seen that even at a depth of 20 cm the depth dose has only fallen to some 55% of the maximum. The flat type of isodose curve results from the use of the copper filter, which reduces intensity along the beam axis.

For larger fields the distribution is essentially the same as given in Fig. 7 but, the isodose curves extend over a larger area. The percentage depth doses along the axis of the beam are the same regardless of the size of the field (4). In Fig. 8 is shown the distribution of radiation for a 6-cm circular field using 400-kv radiation with a half-value layer of 4 mm of copper at a focal skin distance of 80 cm. This type of distribution is typical of that which may be obtained with a standard type of therapy machine. It is seen that the maximum dose now occurs at the surface of the body and that the dose falls as one proceeds to greater depths, the dose at 20 cm depth now being only 6% of the skin dose. The advantages of the betatron field of Fig. 7 over that of Fig. 8 are obvious.

The rise in dose with depth, as shown in Fig. 7 for the first few centimeters, is attributable to the method of interaction of the high-energy X-ray beam with a scattering medium. At high energies Compton electrons and electron pairs are projected primarily in the forward direction when X-ray photons interact with the scattering



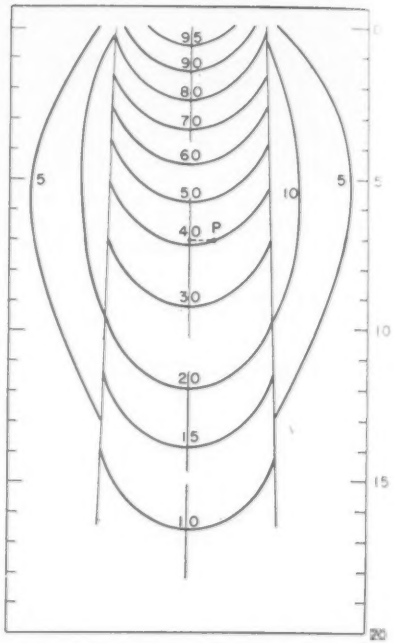


FIG. 7 (left). Isodose distribution for a 6-cm circular field and a focal skin distance of 105 cm, using radiation from a 22-Mev betatron with a copper compensating filter

FIG. 8 (above). Isodose distribution for a 5-cm circular field using 400-kv radiation with half-value layer of 4 mm of copper at 80 cm focal skin distance

medium. These high-energy electrons have ranges amounting to several centimeters in tissue. As one proceeds below the surface of the tissue, the number of secondary electrons tracks increases until equilibrium is obtained, after which point the dose decreases in an exponential manner corresponding to the absorption of the primary radiation. It has been shown (4) that the variation of depth dose with depth has the form $e^{-\mu_1 x} - e^{-\mu_2 x}$, where μ_1 and μ_2 are the linear absorption coefficients of the primary and secondary radiation.

In cancer therapy one is primarily interested in using radiation fields in such a way that the radiation is concentrated at the tumor and no radiation is absorbed in the surrounding healthy tissue. This ideal can never be achieved in practice but can be more nearly approached with a high-energy X-ray machine such as the betatron than with a standard therapy machine.

In Figs. 9 and 10 are shown the resultant distributions obtainable when four fields are combined as indicated, in the treatment of stage four cancer of the cervix. Fig. 9 shows the resultant distribution when 400-kv radiation is used.

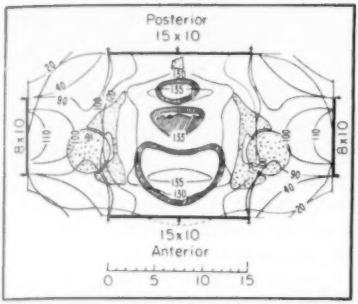


FIG. 9. Isodose distribution for 400-kv radiation

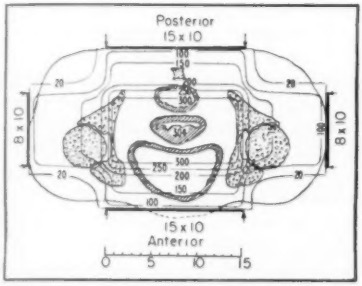


FIG. 10. Isodose distribution for 22-Mev radiation

Where the four fields overlap a maximum dose of 133% is obtained. However, this dose is not much greater than that received by other parts of the pelvis.

In Fig. 10 the distribution for betatron fields is shown. Here a dose of 304% is obtained where the four fields intersect. The rest of the pelvis receives a dose less than ½ of this.

It is obvious that with the betatron it is possible to concentrate the radiation more in the region of the tumor. When 22-Mev radiation is compared with 400-kv radiation in exactly similar arrangements of fields, for the same tumor dose in both cases (*5*), the amount of energy absorbed by the patient using high-energy radiation is between ½ and ⅓ of that received using a standard therapy machine. The two distributions given here should illustrate the physical advantages of betatron radiation over radiation of lower energy.

It is too early to make definite statements concerning clinical results, but preliminary results of the use of the betatron in this way appear encouraging. Clinical experience indicates that patients can tolerate courses of treatment with the betatron that would be impossible with a standard therapy machine. In ordinary therapy the dose received by the skin is often the factor which limits the amount of radiation which can be given a tumor. With the betatron this is never the case, because for a single field the skin dose is only about 10% of the maximum. The relatively small energy absorption in the patient for a given tumor dose is also a great advantage.

Conclusions

Experience with a 22-Mev betatron over the last 18 months indicates that from a physical point of view the betatron is a practical tool in the treatment of deep-seated cancers. Skin reactions are avoided. The distributions of radiation are better than can be achieved at lower energies. The output of the betatron is high enough to make treatment times conveniently short.

The sound which accompanies the operation of the betatron is a disadvantage but does not appear to seriously worry the patient. Further clinical work is necessary before the true potentialities of this machine may be established.

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3. L. Schiff, private communication
4. H. E. Johns *et al.*, *Am. J. Roentgenol. Radium Therapy* **62**, 257 (1949)
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Nuclear Engineering Course at New York University

Last month, NYU, for the second time, offered enrollees its one-year graduate course in Nuclear Engineering. Judging by the interest industrial organizations showed last year in subsidizing employee attendance, the faculty, staffed by NYU and AEC personnel, this year look forward to even greater company participation and eventual cooperative research. Prof. FRANK D. MASLAN, who, assisted by special lecturers, teaches most of the course, has written this commentary on its development

UNRESTRICTED DOCUMENT
DECLASSIFIED BY AUTHORITY OF THE DOE CLASSIFICATION OFFICE

In September, 1949, the College of Engineering of New York University started a graduate course in nuclear engineering administered by the Chemical Engineering Department. A one-year course, it aims to provide an introduction to the whole field of nuclear engineering.

It was organized to supply the present need for all kinds of engineers having an understanding of the fundamentals and applications of nuclear engineering, and was designed to suit the requirements both of engineers active in the field and of those looking forward to such work. To accommodate the many kinds of technical personnel needed in this field, the university decided to admit engineers, physicists and chemists with baccalaureate degrees.

Many leaders in the nucleonics field, both in the Atomic Energy Commission and in private industry, have pointed out that further rapid advance in the field of nuclear technology depends on the dissemination of knowledge among, first, the working technical population of the country and, second, those who expect to take their places in this group. As an example, the Industrial Advisory Group of the AEC* concluded that large industrial participation cannot be expected without much more widespread knowledge of nuclear technology among industrial executives, engineers and scientists.

What comprises Nuclear Engineering was the first question planners of this course had to attempt to answer. As far as an engineer is concerned, nuclear engineering may be divided into two categories: first, the extraction and preparation of suitable materials for nuclear reactors and, second, the design and operation of the reactors themselves. The first division is commonly called process engineering and is related to operations occurring daily in various other industries. Most of this comes within the realm of the chemical engineer. The second division is the new subject matter in nuclear technology. It represents a field of physics which did not exist to any extent prior to 1940. In scope, both financially and in total number of employees, the design and operation of nuclear reactors is smaller than the other processes connected with the field.

Inasmuch as this was among the first attempts of any university to offer such a course, the preliminaries to selecting the subject matter of the course called for considerable discussion with leaders of nuclear technology both in and outside the government during the summer of 1949. Two broad possibilities for a declassified nuclear engineering course appeared to offer themselves: first, a good survey of the field

* Report of the Industrial Advisory Group of the Atomic Energy Commission (released by U. S. Atomic Energy Commission, Dec., 1948).

with emphasis on the engineering technology in all aspects; and, second, a thorough grounding in the theoretical fundamentals of the physics, chemistry and mathematics used in nuclear technology. These two ways of training men are geared to slightly different time sequences. Those people concerned with pushing practical application of nuclear technology now were, of course, interested in a survey-type course for practicing engineers, while those people concerned more with the fundamentals and the long-term development were naturally interested in the second teaching method.

It is obvious that extensive coverage in both of these categories could not be given in one course. For this reason, NYU decided that its best solution of this curriculum problem was to offer a course on the graduate level and make it a survey course together with an introduction to the fundamental physics and chemistry. Such a course, it seemed, would answer both immediate needs of the field and serve as an eye-opener to those students who might wish to pursue the subject matter more intensively. This solution represented a compromise between the two main points of view.

Course Outline

The course, constructed to meet the above requirements, is outlined on pages 86–87.

There are two class sections, one afternoon and the other evening. Each meets once a week for a lecture of two hours and five minutes.

Obtaining Information

Probably the biggest obstacle an educator faces in this field is that of obtaining suitable declassified material for the course. If he wishes to obtain information, it can be had, but largely he must go out and dig it up himself. The sources are many, and there is a great deal of information available.

Generally speaking, AEC people give excellent cooperation and are pleased to help out in an educational endeavor of this type. Securing up-to-date material for this course involves a continuous search of the scientific literature in engineering, physics and chemistry, the perusal of literature in the AEC national laboratories' libraries and public documents issued by the AEC and other government agencies and contractors (e.g., Office of Naval Research contractors), and, not to be underestimated, personal discussion with various experts in the field.

A partial list of general references and the textbooks for this course are given in the appendix on page 87.

Students

Every field of engineering, including administrative engineering, physics, and chemistry, was represented among the forty-five students who took the course last year. Their backgrounds were quite varied. In the day section, most were graduate students, with little or no industrial experience. The evening section comprised mainly older men, most of whom were employed as engineers in various industrial companies in the New York area. Some of them were experts in such fields as heat transfer and distillation, and those who were actively engaged in engineering-design work on nuclear processing found the course valuable background for increasing their understanding of the whole field.

Companies' Interest

Some companies permitted their employees time off to attend the course; others saw fit to pay their employee's tuition. This was not entirely unexpected inasmuch as discussions with representatives of many companies had shown that they were interested in the nucleonics field, but were hesitant to

NUCLEAR ENGINEERING COURSE OUTLINE—New York University

FIRST TERM

1. Introduction
- a) History of atomic energy
- b) Brief description of the U. S. Atomic Energy Commission
- c) Geology, geography and production of raw materials for nuclear reactions

Fundamentals

2–6. Nuclear Physics for Engineers—brief introduction to nuclear physics
- a) Theory of elements, atoms, molecules and isotopes
- b) Atomic and nuclear particles
- c) Detection of nuclear particles
- d) Methods of accelerating atomic particles—brief
- e) Nuclear reactions
- f) Transmutation
- g) Radioactivity
- h) Nuclear fission
- i) Units—definitions and conversions

7–8. Chemistry
- a) Radiochemistry
- b) Chemistry of the heavy elements
- c) Transuranium elements
- d) The chemistry of the fission process

Nuclear Reactors

9-11. Elementary Pile Theory and Application
- a) Cross sections and absorption
- b) Diffusion theory
- c) Pile equations
- d) Critical pile dimensions
- e) Reflectors
- f) Control and operation of a pile

12. Design and Construction of Nuclear Reactors
- a) Graphite pile
- b) Heavy water pile
- c) Breeding
- d) Shielding of reactors
- e) Description of Canadian National Research Council low-power nuclear reactor at Chalk River, Ontario

13–14. Heat Transfer Problems in Nuclear Reactors
- a) Coolants—general
- b) Liquid metals
- c) Suspensions of metals in liquids
- d) Steady-state heat transfer design methods
- e) Unsteady-state heat transfer design methods

14.5. Power Production

Discussion of methods and economics for utilizing energy developed by nuclear reactors. The place of nuclear power in the general utilities complex. This takes last part of Lecture 14.

15. Health and Safety Engineering
- a) Hazard evaluation
- b) Tolerance standards
- c) Instrumentation
- d) Shielding for personnel safety
- e) Radiation sickness

SECOND TERM

16. Disposal of Radioactive Wastes
- a) Radioactive solids
- b) Radioactive liquids
- c) Radioactive gases

Materials

17. Action of Radiation on Materials
- a) Gases
- b) Liquids
- c) Solids
- d) Organic and inorganic materials
- e) Living organisms
- f) Catalysis of chemical reactions

18. Materials of Construction
- a) Metallurgy
- b) Piles
- c) Chemical process equipment
- d) Corrosion

Separation Processes

19. Theory of Cascade Processing

Application to physical and chemical separations.

20. Gaseous Diffusion

Discussion and engineering of separation of isotopes by diffusion through membranes.

21. Thermal Diffusion
- a) Theory
- b) Application to separation of isotopes

22. Ion Exchange
- a) Theory
- b) Engineering design methods
- c) Separation of isotopes
- d) Separation of rare earths

23. Other Separation Methods—*I*
- a) Solvent extraction, uranium purification and separation
- b) Gaseous absorption, recovery of uranium hexafluoride
- c) Adsorption

(Lecture Nos. are shown in boldface type)

24. Other Separation Methods—*II*
- a) Preparation of heavy water
- b) Chemical separation methods, plutonium
- c) Concentration of isotopes by the electromagnetic method

25. Vacuum Technology in Nuclear Engineering
- a) Pumps
- b) Fluid Flow
- c) Measuring devices
- d) Leak detection
- e) Applications of high vacuum technology

26. Instrumentation
- a) Radiation measurements
- b) Pile control
- c) Remote control operations, maintenance
- d) Devices for handling radioactive materials

Applications of Nuclear Engineering

27. Radioisotopes
- a) Production
- b) Medical applications
- c) Other scientific applications
- d) Industrial applications

28. Miscellany
- a) Design of radiochemical process buildings and laboratories
- b) Atomic energy for military purposes, bomb and bomb defense
- c) Propulsion of ships

29. Jet Propulsion using Nuclear Energy
- a) Theory
- b) Materials of construction
- c) Cooling methods

30. Social Implications
- a) Description of the U. S. Atomic Energy Commission
- b) Security regulations
- c) Scientific freedom
- d) Civilian vs military control
- e) Federal vs private company enterprise
- f) United Nations international control

enter because of lack of knowledge and trained personnel. The course served to introduce them to the possibilities awaiting development in the field.

Research

One of the corollaries for good instruction on the graduate level in an engineering course should be a coordinated research program. Such a program is under way in our College of Engineering. Work at present is going on in cosmic-ray measurements, Geiger counter development, heat transfer, waste disposal, and instrumentation.

Future Developments

Future plans call for progress along two coordinated channels. As the field of nuclear engineering develops, more courses will be offered. Class work will, in turn, be strengthened by fundamental engineering research and by cooperative research in conjunction with interested companies.

APPENDIX

General References for the Course

W. Blaedel, "Nuclear Chemistry" (NEPA Division, AEC, Oak Ridge, Tenn.)

C. D. Coryell, editor, "An Introduction to Nuclear Chemistry, Lecture Series" (U. S. Atomic Energy Commission MDDC-763)

G. Friedlander, J. W. Kennedy, "Introduction to Radiochemistry" (John Wiley & Sons, Inc., New York, 1949)

C. Goodman, editor, "The Science and Engineering of Nuclear Power," Vols. I and II (Addison-Wesley Press Inc., Cambridge, Mass., 1947) *

Hanford Engineer Works, "Nuclear Physics Primer," Chapter 1 (U. S. Atomic Energy Commission MDDC-1497)

C. Helmholz, "Course in Nuclear Physics for Engineers" (U.S. Atomic Energy Commission MDDC-1014)

O. Oldenburg, "Introduction to Atomic Physics" (McGraw-Hill Book Co., Inc., New York, 1949)

E. Pollard, W. L. Davidson, "Applied Nuclear Physics" (John Wiley & Sons, Inc., New York, 1942)

H. D. Smyth, "Atomic Energy for Military Purposes" (Princeton University Press, Princeton, N. J., 1945)

H. Soodak, E. C. Campbell, "Elementary Pile Theory" (John Wiley & Sons, Inc. New York, 1950) *

W. E. Stephens, "Nuclear Fission and Atomic Energy" (The Science Press, Lancaster, Pa., 1948)

J. Stranathan, "The 'Particles' of Modern Physics" (The Blakiston Company, Philadelphia, Pa., 1942)

* Course textbooks.

BOOKS

The Meaning of Relativity (Third Edition, **including the Generalized Theory of Gravitation**), by Albert Einstein, Princeton University Press, Princeton, N. J., 1950, 150 pages, $2.50. *Reviewed by M. S. Plesset, associate professor of applied mechanics, California Institute of Technology.*

This book, as it first appeared in 1922, consisted of the text of lectures given by Professor Einstein at Princeton University. The topics discussed in the main portion of the book are: Space and Time in Pre-relativity Physics, The Theory of Special Relativity, and The General Theory of Relativity. The discussion throughout the book is truly distinguished for its clarity. The reader who is unfamiliar with relativity theory will be pleasantly surprised with the degree of understanding this text will give him.

An appendix on The Cosmologic Problem was added to this book when the second edition appeared. In the period between the first and second editions, the important discovery was made of the shift toward the red end of the spectrum of the spectral lines from extra-galactic nebulae. The observations indicated that this red-shift was independent of wavelength so that it was interpreted as a Doppler effect. The data further indicated a velocity of recession of the extra-galactic nebulae which is proportional to the distance. These findings had a profound effect on the theoretical attempts to utilize the approach of general relativity for the cosmological problem of the universe in the large. In this appendix, Einstein gives a review of the theory of the expanding universe.

The third edition of this book has a second appendix on The Generalized Theory of Gravitation. The appearance of this contribution to general relativity was greeted with an amazing furor in press and radio. The scholar may very well be distressed at the garbled accounts given the public of an abstruse analysis of a problem in theoretical physics, but the popular interest certainly exists. The problem discussed in this appendix is one which Einstein has been considering for some time; it concerns an extension or "unification" of general relativity.

The field equations of general relativity relate the fundamental metric tensor g_{ij}, and quantities derived from it, to the energy tensor of matter T_{ij}. As Einstein expresses it, "T_{ij} represents the energy which generates the gravitational field, but is itself of non-gravitational character, as for example the energy of the electromagnetic field, of the density of ponderable matter, etc. To represent this tensor, T_{ij}, concepts are taken from pre-relativity physics which only *à posteriori* have been adapted to the principle of general relativity." This dualism appears conceptually unsatisfactory to Einstein.

In the extension of general relativity proposed here, Einstein accepts a drastic modification of conventional Riemannian geometry. In Riemannian geometry, the metric tensor g_{ij} is always symmetric in its indices. Einstein proposes that the tensor g_{ij} be allowed to have an anti-symmetric as well as a symmetric part. The Riemann-Christoffel symbols, which also in Riemannian geometry have definite symmetry properties, no longer have these properties. A generalization of this kind has been considered by mathematicians. When the Reimann-Christoffel symbols have the usual symmetry, the space has curvature; if the symmetry is not required, the space will also possess torsion. The field equations in a space so generalized can include, as Einstein shows, not only gravitational equations

but the equations of electrodynamics as well. The mathematical difficulties so far have prevented a determination of the physical consequences of the theory so that it is not yet possible to assess the success of the theory.

While the appendices may be of more specialized nature, the text itself can be highly recommended to the engineer or scientist who wishes to get a general view of relativity; the reader who has a familiarity with the theory will also derive great pleasure from this book.

Biological Studies with Polonium, Radium, and Plutonium (Div. VI, Vol. 3, of National Nuclear Energy Series), edited by Robert M. Fink, McGraw-Hill Book Co., Inc., New York, 1950, xvi + 411 pages, $3.75. *Reviewed by Austin M. Brues, M.D., director of the Division of Biological and Medical Research, Argonne National Laboratory.*

This 400-page monograph is a distillate of the studies made on three important radioactive elements by the biological research group at the University of Rochester for the Manhattan Project and the Atomic Energy Commission.

It is divided into three parts: The first part has to do with the distribution and excretion of polonium and involves animal and human polonium metabolism. The second part is on the distribution and excretion of radium in rats, and includes a valuable review of the literature in radium toxicity and metabolism by H. G. Silverstein. The final part gives data on the toxicities of radium, polonium, and plutonium in rats, including a discussion by G. A. Boyd and R. M. Fink of the relative toxicities of the three elements in terms of total alpha-particle energy dissipated in the body.

It is obvious that, on this physical basis, polonium and plutonium are much more toxic than radium, and that the discrepancy is due to the tissue distribution of the elements, but this has not yet been translated into terms of tissue radiosensitivities to an extent that makes it possible to predict other radiotoxicities.

Also, unfortunately, the bulk of the survival data covers only a period of a few months after irradiation; further work will be required to establish the toxicities of polonium and plutonium relative to radium, which alone has a human history of chronic toxicity. The data serve to emphasize that the alpha-emitting elements are remarkably toxic, yet it has been demonstrated that these elements can be safely handled in large amounts if proper precautions are followed.

As noted in a preface by the Editorial Advisory Board, the preparation of a unified account of atomic energy project work suffered from the effects of the rigorous compartmentation applied during the war. This complicated somewhat the task of this board. The existence of other simultaneous work on the heavy radioelements did not, however, involve serious duplication, but only the sort of duplication which, as in the free scientific society, stimulates further thought and work. The policy of this board is such that it has been possible to publish in extraordinary detail the protocols and conclusions of an important pioneering series of experiments.

The Liquid Metals Handbook, R. N. Lyon, editor-in-chief, Superintendent of Documents, U. S. Govt. Printing Office, Washington 25, D. C., 1950, viii + 188 pages, $1.25. *Reviewed by E. R. Gilliland, Dept. of Chemical Engineering, Massachusetts Institute of Technology.*

This handbook represents the work of a number of authors who collaborated under the aegis of the Office of Naval Research, the Atomic Energy Commis-

sion, and the Bureau of Ships. It represents a compilation of the published literature, plus information obtained from reports of the Atomic Energy Commission and the Navy Department. It also includes material based on personal communications from a number of investigators who have worked with liquid-metals.

Such fields are covered as industrial utilization, physical and chemical properties, laboratory techniques, corrosion problems, availability, heat transfer, and the practical handling of the materials on an industrial scale. The material is well organized and complete.

The book covers, as liquid-metals, most of the metallic elements with melting points below 700° C as well as some of their alloys. It was undoubtedly prepared because of the interest in liquid-metals as coolants for nuclear reactors. However, *anyone* working with these metals will find the information given in this book to be helpful. In the past it has been difficult to obtain information on these materials due to the fact that the data on most of them have been widely scattered in the literature and a large part of them were in reports that were not readily available. The handbook performs a real service in bringing this information together and in evaluating it.

BOOKS RECEIVED

Counting Tubes—Theory and Applications, by S. C. Curran and J. D. Craggs, Academic Press, Inc., New York, 1949, xi + 238 pages, $5.50.

Clinical Use of Radioactive Isotopes, by Bertram V. A. Low-Beer, Charles C Thomas, Springfield, Ill., 1950 xv + 414 pages, $9.50.

Principles of Nuclear Chemistry, by Russell R. Williams, Jr., D. Van Nostrand Company, Inc., New York, 1950, ix + 307 pages, $3.75.

The Philosophy of Mathematics, by Edward A. Maziarz, Philosophical Library, New York, 1950, viii + 286 pages, $4.

Unit Operations, by George Granger Brown *et al.*, John Wiley & Sons, Inc., New York, 1950, xii + 611 pages, $7.50.

The Theory and Practice of Industrial Research, by David Bendel Hertz, McGraw-Hill Book Company, Inc., New York, 1950, xiii + 385 pages, $5.50.

The Human Use of Human Beings, by Norbert Wiener, Houghton Mifflin Company, Boston, Mass., 1950, 241 pages, $3.

Secret, by Michael Amrine, Houghton Mifflin Company, Boston, Mass., 1950, 311 pages, $3.

The Evolution of Scientific Thought from Newton to Einstein (second edition), by A. d'Abro. This is an enlargement and a revision of the first edition published in 1927. The chapter on the finiteness of the Universe has been rewritten entirely and now includes a brief discussion of the Expanding Universe of the Abbé Lemaitre. Dover Publications, Inc., New York, 1950, xx + 481 pages, $3.95.

OTHER LITERATURE

Nuclear Data, National Bureau of Standards Circular 499. A tabular compilation of experimental values of half-lives, radiation energies, relative isotopic abundances, nuclear moments, and cross sections. Decay schemes and level diagrams are provided wherever possible. At present over 1,000 new measurements of nuclear properties are being reported each year; supplements to the present loose-leaf compilation will be issued at six-month intervals to keep it up to date. *Available from Superintendent of Documents, U. S. Govt. Printing Office, Washington 25, D. C., $4.25 including three supplements.*

Chemical Economics Handbook. This is a new type of encyclopedic information service designed for executives and others planning company economic policies. During 1950, 250 chemical economic data sheets will be issued. Only new data sheets will be issued during 1950–51; during subsequent years, revised sheets will be issued along with additional new ones. *Available from the Stanford Research Institute, Stanford, Calif.; subscription price, $250 per year; each additional concurrent subscription to the same organization, $60 per year.*

German-English Technical Dictionary, Vol. I, A–K. Based on data compiled by U. S. Air Force, edited by Kurt F. Leidecker. The over 100,000 terms it contains were collected in connection with the job of screening and processing the tons of air-technical information captured from Germany after World War II. Of particular use to readers of NUCLEONICS may be the terms included under, to name a few, the subjects of rocketry, atomic physics, higher mathematics, electronics, and infrared research. Vol. I is available now; Vol. II, L–Z, will be available later this month. S. F. Vanni, 30 W. 12th St., New York 11, 1950, both volumes $35.

United Nations, International Control of Atomic Energy: Official Records, Fourth Session, Supplement No. 15. *Available from Columbia University Press, New York 27, N. Y., $0.30.*

Management of Industrial Research (A Selected and Annotated Bibliography). *Available from Arthur D. Little, Inc., Cambridge 42, Mass., no charge.*

Attempts to Prevent and Counteract Effects of Ionizing Radiations by Chemical and Pharmacological Means, (NEPA-1127) by W. A. Selle. *Available from Office of Technical Services, Dept. of Commerce, Washington 25, D. C.*

The Development of a Policy for Industrial Peace in Atomic Energy (Planning Pamphlet No. 71, 104 pages), by Donald B. Straus, executive secretary of the Atomic Energy Labor Relations Panel. *Available from the National Planning Association, 800 21st St., N.W., Washington 6, D. C., $1.*

The Hydrogen Bomb and International Control: Technical and Background Information. Pamphlet 65616, July, 1950, 41 pages, printed for use of the Joint Committee on Atomic Energy, 81st Congress, 2d Session.

Methuen Monographs on Physical Subjects, available from John Wiley & Sons, Inc., New York: *Wave Guides* (3d ed.), by H. R. Lamont, $1.50; *Electromagnetic Waves* (4th ed.), by F. W. G. White, $1.25; *The Special Theory of Relativity* (3d ed.), by H. Dingle, $1.25.

British Declassified Reports on Atomic Energy. The 61 new reports (BDDA Nos. 177–237) that were recently made available for sale, supplementing those previously listed in NUCLEONICS (Jan. '48, p. 65; Mar. '48, p. 37; June '49, p. 71), are listed in Ministry of Supply Release DS 42847, dated July 14, 1950. The reports themselves may be ordered in the form of enlarged prints (9 by 7 inches) from microfilms from H. M. Stationery Office, P. O. Box 569, London, S.E.1.

British Manual on Atomic Warfare. *Available from British Information Services, 30 Rockefeller Plaza, New York 20, N. Y., $0.50.*

Indium. This book is said to be the first complete and authoritative compilation of material on indium, its occurrence, metallurgical and chemical characteristics, and history since its discovery in 1863. *Available from The Indium Corporation of America, 60 East 42nd Street, New York 17, N. Y., $7.50.*

CROSS SECTIONS

A Scintillation Geiger Counter*

By C. E. Mandeville

Bartol Research Foundation of the Franklin Institute
Swarthmore, Pennsylvania

Nuclear counting has been greatly facilitated in recent years by the development of scintillation techniques. Very high efficiencies for gamma rays have been obtained when scintillations from gamma-irradiated crystals have been detected by commercially available photomultipliers. Rapid progress has been made. It would have been difficult to predict several years ago the use of photomultipliers in coincidence spectrometers which today determine the quantum energies of gamma rays. For some purposes, it has appeared that crystals used in conjunction with photomultipliers would replace the time-honored Geiger-Müller counter. It is well known that the photomultiplier is free of certain objections to Geiger-counter operation, such as the relatively long "dead time" and the finite life of the Geiger counter. Of course, these two particular failings of Geiger counters can be circumvented by the use of such tubes in the proportional region. There remains, however, the problem of detection efficiency, and, particularly for gamma detection, it has seemed that the photomultiplier scintillation counter holds many advantages.

Since 1947, there has remained in the minds of the members of this laboratory the thought that perhaps Geiger counters themselves could be used for the detection of scintillations excited when crystals are irradiated by nuclear particles. This idea was born of the fact that photon counters, light-sensitive counters, had long been in use at the Bartol Research Foundation. Counters of this type were highly developed by Gordon Locher at Bartol.

In the course of Geiger counter investigations at Bartol, a method of counter sensitization involving a discharge treatment at liquid-air temperatures was discovered by Scherb (*1*).

Over a period of time, attempts were made to reproduce the work of Locher in obtaining counters of high sensitivity, and counters were sensitized according to the method described by Scherb. But no response was obtained in these counters when currently available scintillating crystals were irradiated by nuclear particles. The negative result arose from the fact that the spectral response of the counters did not match the frequencies of the radiations emitted by the crystals. Finally, it was decided to concentrate primarily upon the development of crystals which would fluoresce in the ultraviolet region and thus actuate the available Geiger counters having spectral responses peaked in this region.

Glasser and Beaseley (*2*) describe experiments wherein they observed a long period phosphorescence or "after-

* The development of this instrument was assisted by the joint program of the Office of Naval Research and the Atomic Energy Commission.

glow" in a photosensitive Geiger counter when they irradiated crystals of NaCl with gamma rays of radium. This report served as a starting point for measurements at this laboratory in which NaCl, activated by such metals as thallium and silver, was irradiated by nuclear particles. Immediate fluorescent pulses in the ultraviolet were detected in a photosensitive Geiger counter when crystals of NaCl-Ag were irradiated by alpha particles. By the use of Scherb's method of sensitization, a very high quantum efficiency was attained. It finally became possible to detect short-lived ultraviolet scintillations from both beta rays and gamma rays on NaCl-Ag. By surrounding a photosensitive counter with crystals of NaCl-Ag, it has been observed that the detection efficiency of the counter for gamma rays is greatly increased.

It has been found that several other crystals emit ultraviolet detectable in the Geiger counter when irradiated by alpha and beta rays. These are NaBr-Ag, NaBr-Tl, KCl-Ag, LiCl-Ag, and LiBr-Ag. Thus far, only NaCl-Ag has responded sufficiently to gamma rays to yield enough ultraviolet photons for detection in photosensitive Geiger counters.

It seems that the scintillation Geiger counter would have particular use in the field as an alpha detector for decontamination purposes. Such an instrument would employ only a "light-tight" window. Current alpha detectors are regarded as fragile, because a vacuum-tight thin window is required.

The actual operation of the counter offers the simplicity of the Geiger region in which the individual pulses are large and uniform in size. The proportional region of the scintillation Geiger counter is as yet uninvestigated.

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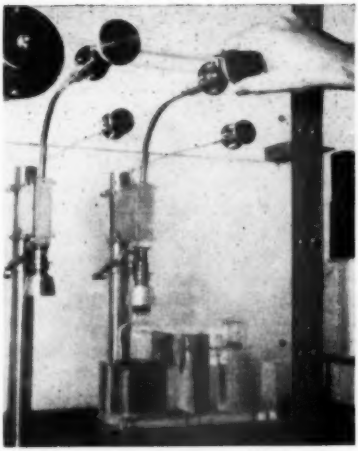
1. M. V. Scherb, *Phys. Rev.* **73,** 86 (1948)
2. O. Glasser and I. E. Beasley, *Phys. Rev.* **47,** 789 (1935); *Phys. Rev.* **47,** 570 (1935)



Remote Decapping Device Used to Handle Isotopes

Designed by the engineering department of Oak Ridge National Laboratory, the decapping device shown here simplifies the handling of radioisotopes. With this device it is possible to uncap and cap bottles of radioactive materials at distance as great as 10 feet behind thick barriers.

The bottle cap is grasped by a hollow rubber cone when the knob of the flexible cable is pushed in. The knob is then turned to unscrew the bottle cap. Pulling the knob removes the cap from the bottle.

When replacing the cap on the bottle, the same operations are carried out in reverse order, with the exception that a lever is actuated by a pull cable to break the grip of the cone on the capped bottle. A rubber pad is used under the bottle to keep it from turning.

Labeled Alcohol, Deuterium Used to Study Fuel Catalysts

Radioactivity measurements have been used to clarify two aspects of the catalytic mechanism by which coal is converted to gasoline. Labeled alcohol

has been used to show that alcohol molecules adhere to the catalyst surface and serve as starting points for the building of hydrocarbon molecules. The use of deuterium permits the measurement of the rate at which a catalyst wears out in use.

J. T. Kummer, H. H. Podgurski, W. B. Spencer, and P. H. Emmett of the Mellon Institute of Industrial Research, Pittsburgh, Pa., found that hydrocarbons formed in the process when radioactive alcohol was used, had the same activity per volume regardless of the number of carbon atoms in each molecule. This indicates that alcohol molecules can serve as the starting point of the synthesis.

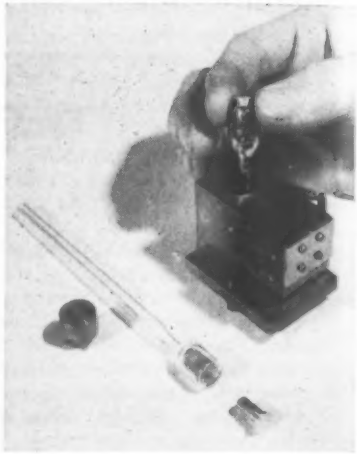
Analysis of the rate of deterioration of the catalyst in the Fischer-Tropsch process requires measurement of the carbon present as iron carbide. Using deuterium in the conversion of this carbon into a hydrocarbon makes it possible to separate it from gasoline-synthesis hydrocarbons by mass spectrometry. The method has been used by H. I. Weck, S. Meyerson, and H. S. Seeling of the Standard Oil Co. of Indiana, Whiting, Indiana, to obtain data useful in catalyst development.

These experiments were reported at the 118th national meeting of the American Chemical Society, Chicago, Ill., on September 6.

Utilize Differential Expansion To Open Ampoule Remotely

The difference in the thermal expansivity of aluminum and glass is utilized in technique developed by Jesse Sherwood of the National Bureau of Standards for introducing highly reactive materials into a closed system.

The material to be introduced—in this case cesium—is distilled into a specially designed ampoule and sealed off under vacuum. The ampoule, as shown in the illustration, contains a



well in which a carefully lapped aluminum plug of about 7 mm diameter is inserted.

After the ampoule is filled and sealed, the aluminum plug is inserted. The assembly is then placed in the system into which the material is to be introduced. When the ampoule is heated to 80–100° C, the aluminum plug, expanding more than the glass, will crack the glass and release the contained compound.

Plutonium Produced by Fission in Pitchblende

Microgram quantities of Pu^{239} have been obtained from the liquid wastes resulting from the extraction of uranium from Belgian Congo pitchblende concentrate. The sample's identity was proved by a study of its chemical, radioactivity and fission characteristics. The Pu^{238} content was shown to be less than 0.0003% of the Pu^{239} content. Plutonium isotopes with masses greater than 239 were found to be less than 1% as abundant as Pu^{239}.

In reporting their work at the 118th national meeting of the American Chemical Society, Chicago, Ill., on

September 4, Dr. D. F. Peppard and his associates suggested that the ore deposit behaves like a man-made uranium pile, producing Pu^{239} by fission and neutron absorption by U^{238}. The authors pointed out, however, that the possibility exists that part of the Pu^{239} could be present as a daughter of a hitherto undiscovered, naturally occurring radioactive series.

IN BRIEF

Tritium has been found in ordinary water by W. F. Libby, University of Chicago, and A. Von Grosse, Temple University, in the ratio of one atom of tritium to every 10^{18} atoms of normal hydrogen.

Silicone coating on glass apparatus minimizes adsorption of P^{32}, a source of error in tracer experiments. B. A. Rubin, Brookhaven National Laboratory, has found that the coating also makes decontamination easier.

Slide rule for calculating decay correction factors in activity computations has been devised by J. L. Herson of the National Bureau of Standards.

PATENTS RELEASED

The Atomic Energy Commission has released 23 more patents for nonexclusive, royalty-free use. Copies of the patents listed can be obtained from the U. S. Patent Office, Washington, D. C. For licenses, apply to the Chief, Patent Branch, Office of the General Council, U. S. Atomic Energy Commission, Washington, D. C.

Detecting Device (2,513,805), W. R. Kanne. Inexpensive fluorescent instrument utilizes contrasting visual effects to detect radioactivity.

Method of Dissolving Difficultly Soluble Metal Sulfates (2,514,115), A. H. Angerman.

Induction Reamer (2,514,116), W. R. Baker. Unattached circular tool is rotated inside tube by means of rotating magnetic field.

Radiation Detector (2,514,135), H. G. Neil. An audible note distinguishes intensity and frequency of radiation.

Hydraulically Driven Oscillatory Brush (2,514,142), J. D. Reid. Brush for cleaning can be held in fluid-tight contact with surface being cleaned.

Method for Disposing of Corrosive Gases (2,515,112), W. B. Burford, III, H. C. Anderson. Caustic acid solution with sodium sulfide is used to remove fluorine.

Quick Operating Valve (2,515,159), E. Zurcher.

Adjustable Centrifugal Switch (2,516,-050), Q. J. Evans, A. Kushner.

Method and Apparatus for Measuring Alpha Particle Radiation (2,517,469), R. W. Dodson, W. H. Beamer. Neutrons from (α,n) reaction are measured.

Pulse-Forming Preamplifier (2,517,676), Q. A. Kerns.

Voltage Supply Circuit for Vacuum Tubes (2,517,863), D. K. Froman.

Apparatus for Testing Tensile Characteristics of a Material (2,518,217), C. K. Beck, D. Kirkpatrick. Test piece is maintained at constant temperature.

Radiation Counters (2,519,007), V. C. Wilson. Instrument for measuring fast neutrons.

Manufacture of Oxygen-containing Chlorinated Cyclic Compound (2,519,319), E. T. McBee, J. S. Newcomer. Method for making chloranil.

Method of Measuring Corrosion and Erosion (2,519,323), W. B. Shank, E. C. Creutz, D. H. Gurinsky.

The Electrolytic Production of Metallic Uranium (2,519,792), R. Rosen.

Method and Apparatus for Separating Fluids by Thermal Diffusion (2,521,112), J. W. Beams.

Dispersion Separation (2,521,121), M. Kilpatrick.

Secondary Electron Multipliers (2,521,-133), A. H. Snell, L. C. Miller.

Carbonaceous Articles and Production Thereof (2,521,495), H. A. Wilhelm, P. S. Gerald.

Process for Fluorinating Hydrocarbons (2,521,626), A. F. Benning. Petroleum lubricating oil fraction is fluorinated.

Acoustic Chamber for Analysis of Gaseous Mixture (2,521,634), W. H. Janssen, W. Mikelson. Effective molecular weight is determined by measuring natural frequency of chamber filled with gas.

Ionization Chamber (2,521,656), E. G. Segre, O. Chamberlain. Hydrogenous material supports metal portions of chamber and fills interior, slowing fast neutrons to thermal energies.

NUCLEONIC EVENTS

TRUMAN APPROVES INCREASE IN AEC SALARIES

Last month in Congress a resolution was passed to raise the salary of the chairman of the Atomic Energy Commission from $17,500 to $20,000 a year and that of the other commission members from $15,000 to $18,000. The resolution, which also increases the salary of the general manager from $15,000 to a permissible maximum of $20,000 annually, further provides that the AEC will appoint the general manager rather than the President with the advice and consent of the Senate, as is the present practice.

President Truman approved the legislation on September 24.

AEC ANNOUNCES RULES FOR CONTRACT APPEALS

The Atomic Energy Commission recently announced the "Rules of Procedure" of its Advisory Board of Contract Appeals. The Board was established last February to hear appeals originating under the "disputes article" of AEC contracts and to make recommendations to the General Manager of the Commission for their disposition.

The disputes article provides that contractors may appeal the decisions of AEC contracting officers in disputes arising under the terms of their contracts with the AEC. The final decision on each appeal is made by the General Manager on the basis of recommendations submitted by members of the Advisory Board.

The Rules of Procedure, which were developed by the Advisory Board in consultation with the Commission's staff and formally approved by the Commission, were published in the August 30 edition of the *Federal Register*.

ORINS OFFERS THREE MORE RADIOISOTOPE COURSES

The eighteenth, nineteenth, and twentieth courses in the techniques of using radioisotopes in research will be given by the Special Training Division of the Oak Ridge Institute of Nuclear Studies during the winter and spring of 1951. Dates for the courses are as follows:

January 8–February 2
February 19–March 16
April 16–May 11

The Special Training Division can accommodate thirty-two participants at each of the three courses. A registration fee of $25 is charged, and participants will bear their own living and traveling expenses.

Additional information and application blanks may be obtained from Dr. Ralph T. Overman, Chairman, Special Training Division, Oak Ridge Institute of Nuclear Studies, P. O. Box 117, Oak Ridge, Tennessee.

PATENT HOLDER SUES U. S., ASKS PAY FOR FERMI PROCESS

On August 21, an attorney for G. M. Giannini & Co., Inc., Pasadena, Calif., the firm that holds patent rights to a process developed by Enrico Fermi and co-workers and used in atom bomb manufacture, filed a $10,000,000 damage suit against the Government and charged use of the process without payment.

This suit was separate from another claim filed by the same company with

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the Atomic Energy Commission. The latter claim, for an unspecified amount, is for payment for use of the process since the Atomic Energy Act of August, 1946, which revoked patent rights on the process when it was employed for production of fissionable material.

The suit against the Government asks $7,500,000 for use of the process before the Act and $2,500,000 for its use since that time in the production of radioactive materials for other than military use.

The 1938 Nobel Prize winner, Dr. Enrico Fermi, now at the University of Chicago, developed the process in Rome together with E. Amaldi, Bruno Pontecorvo, Franco Rasetti, and E. Segrè. They were granted a United States patent in 1940 and assigned all rights to Giannini, whom they knew in Rome.

PHEONIX PROJECT AWARDS FUNDS, SPONSORS PILE RESEARCH

Research grants totaling $19,200 have been made by the Preliminary Planning Committee of the University of Michigan's Memorial-Phoenix Project, a $6,500,000 alumni-sponsored program to investigate the implications and applications of atomic energy.

The largest amount, $7,500, was awarded to Fred J. Hodges, chairman of the Department of Roentgenology, to support an isotope laboratory.

H. R. Crane, professor of physics, received a grant of $4,700 to continue construction and operation of a special Geiger counter used in dating 5,000–25,000-year-old relics by means of radiation measurements of carbon-14 atoms.

A grant of $2,600 was awarded to H. H. Bartlett, professor of biology, to support research on human migration through the Aleutian Islands. The study involves the carbon-14 method of dating soil samples from the Islands.

John Lederle, associate professor of political science, was awarded a $1,800 grant to study the activities of Congress in maintaining legislative direction and control over the development of the nation's atomic energy program.

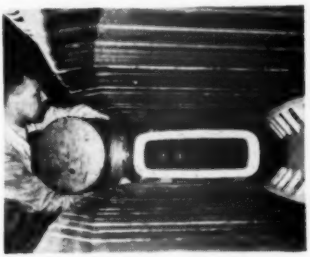
The rate of nuclear energy dissipation of rubidium-87 will be investigated under a grant of $1,200 to Marcellus L. Wiedenbeck, associate professor of physics. Donald A. Glaser, instructor in physics, received a $750 grant for high-energy cosmic-ray research.

Robert W. Parry, assistant professor of chemistry, was awarded a grant of $650 to study ion-exchange resins.

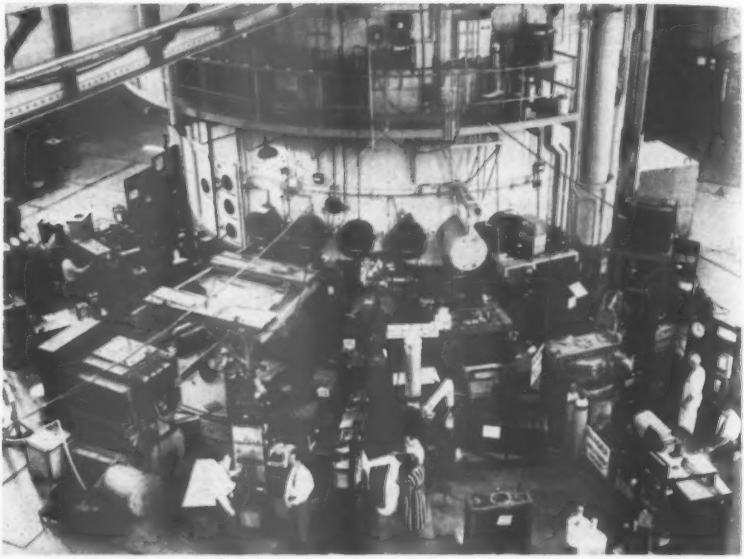
Unclassified Reactor Research

Another recently announced activity of the Phoenix Project will be the support of an investigation of some of the

ABOUT THE COVER



VACUUM box section fits between the magnet pole tips of the proton synchrotron at Birmingham University, England. Pumping ports and a section of the stainless-steel vacuum manifold can be seen. The apparent roughness of the pole faces is caused by the paper insulation between batches of laminations. For a comprehensive description of this accelerator which is expected to be in operation in the near future, see L. U. Hibbard's article "The Birmingham Proton Synchrotron," page 30. Photo from McGraw-Hill World News, London Bureau.



This is the first picture ever released for publication of Canada's heavy-water pile at Chalk River, Ontario. Operated under the direction of the National Research Council, this reactor has produced the highest flux of thermal neutrons ever recorded. Shown in the view above is the congestion of experimental equipment into which the neutron beams pass as it emerges from the various openings in the face of the reactor. Just beyond the young lady in the center is a neutron spectrometer. Above the spectrometer is the apparatus that was used to show the neutron to be unstable and have a half-life of a few minutes. Nuclear energy levels are investigated by means of the special type of electron-pair spectrometer shown at the right

problems implicit in the design and construction of a high-temperature nuclear reactor. The research will be conducted by the University of Michigan's Department of Engineering Mechanics under the direction of Paul F. Chenea and with the assistance of Alfred R. Bobrowsky.

The research proposed is entirely unclassified but liaison on a classified level with the AEC will be maintained to prevent duplication of effort.

RADIOISOTOPE SYMPOSIUM PLANNED BY CHEMICAL SOCIETY

The New York Section of the American Chemical Society has scheduled an all-day "Symposium on Radioisotopes" to be held in the Hotel Statler, New York, on Friday, January 19, 1951. The complete program will be published at a later date.

AEC PLANS RELEASE OF MORE TECHNICAL INFORMATION

The Atomic Energy Commission is making plans to give to industry and the industrial press more technical information in the field of atomic energy.

Norman H. Jacobson, who has been supervisor of Industrial Press Relations for Allis-Chalmers Co., Milwaukee, has been added to the staff of AEC to carry out this program. As Technological Information Officer, he will assist the Chief of Technical Information Service, Dr. Alberto Thompson, in developing policies and programs for dissemina-

tion of such information, valuable to industry.

Specifically, Jacobson's duties will include a continuous appraisal of AEC technological developments; arrangements for proper declassification of such information and its publication where no security angle is involved. He will cooperate closely with the various AEC contractors and the individual scientists.

The appointment of Jacobson comes as the direct result of a recommendation of an AEC industry advisory committee known as the AD Hoc Committee on Technological Information for Industry. The committee, headed by Sidney D. Kirkpatrick, vice president of McGraw-Hill Book Company, and composed of 12 representatives from industry and the industrial press, has been studying for several months ways and means of giving industry more advantages from atomic-energy research (NU, Feb. '50, p. 92).

NBS CREATES BASIC INSTRUMENTATION OFFICE

The National Bureau of Standards has established a new Office of Basic Instrumentation. Created on June 1, 1950, the office coordinates a program of evaluation and improvement of instruments for measuring basic physical quantities.

Plans for Bureau projects in basic instrumentation, maintenance of liaison with sponsoring agencies, and arrangement for cooperative work on special problems will be handled by the office.

NATIONAL RESEARCH COUNCIL FORMS PANEL ON CANCER

The Committee on Growth of the National Research Council recently announced the formation of a Panel on Environmental Cancer. Composed of Drs. Willard Machle (chairman), Francis Heyroth, George H. Gehrmann, Herman Lisko, and Norton Nelson, the panel will evaluate the status of knowledge in this field and formulate criteria for valid relationships between environment and the occurrence of cancer.

The panel will also review applications for grants in support of research in these fields.

WATCH for the

November "NUCLEONICS"

It will feature:

1) A "Special Report on Equipment for Radioactivity Laboratories," containing data classified according to these levels and examples of activity: High (above 1 c)—physical measurements, chemical separations; Intermediate (1 mc to 1 c)—cobalt-60, phosphorus-32; and Low (below 1 mc)—carbon-14, alpha-emitter.

2) NUCLEONICS' Annual "Buyers' Guide" to manufacturers of nuclear instruments, materials, and accessories—a listing that promises to be the most extensive yet published in this field.

NUCLEAR NEWSMAKERS

Franz Newell Devereux Kurie has been appointed as special consultant to the director of research at the Naval Research Laboratory. As superintendent of the nucleonics division, he will plan the nuclear research program.

Eldon C. Shoup, on December 1, resigns as executive vice president of Associated Universities, Inc., the organization that operates Brookhaven National Laboratory for the AEC.

Kenneth H. Kingdon, formerly assistant director of the General Electric Research Laboratory at Schenectady, has been appointed technical manager of the Knolls Atomic Power Laboratory. Other advancements include those of **Lawrence L. Ferguson,** who has been assistant executive engineer, to operations manager of KAPL, and **Walter S. Macaulay,** Rear Admiral, USN(Rtd),

who was also assistant executive engineer, to acting contracts division manager and administrative assistant to KAPL's general manager.

W. Binks of the National Physics Laboratory in Teddington, England, has been appointed secretary of the permanent commission on radiological protection, associated with the International Congress of Radiology.

James M. Cork, professor of physics at the University of Michigan and author of "Radioactivity and Nuclear Physics," recently delivered a series of lectures on nuclear energy to the personnel of General Motors Research Laboratories Division.

The Los Alamos Scientific Laboratory has announced that the following scientists have joined their staff: **John B. Storer,** former research associate at the University of Chicago's Toxicity Laboratory; **Merle E. Bunker,** recently research assistant at Indiana University; **Richard E. von Holdt,** formerly instructor of mathematics at Northwestern University; **Frank C. Hoyt,** former director of Argonne National Laboratory's Theoretical Physics Division; **Alice H. Armstrong,** recently professor at Wellesley College; **William C. Dickenson,** formerly at the Massachusetts Institute of Technology; **Richard L. Henkel,** who had been studying at the University of Wisconsin; **John H. McQueen,** former instructor of physics at the University of Virginia; **Rolf E. Peterson,** formerly a graduate teaching assistant at the University of Wisconsin; **Robert B. Leachman,** who worked at MIT's Radiation Laboratory during World War II; and **Peter D. Lax,** from the staff of the Institute for Mathematics and Mechanics of New York University.

David B. Langmuir, formerly of the staff of the AEC General Manager, has been appointed AEC Liaison Officer at Chalk River to replace **Curtis A. Nelson,** who has been designated manager of the new AEC Operations Office established in connection with the recently announced du Pont contract (NU, Sept. '50, p. 87).

Vincent E. Parker has resigned as chairman of the physics department at the University of Delaware to become head of the department of physics and astronomy at Louisiana State University. He replaces **D. V. Gunthrie,** who has resigned to do full-time teaching in astronomy.

John A. Waters, Rear Admiral, USN (Rtd), has been appointed Director of the Division of Security of the U. S. Atomic Energy Commission. He succeeds **Rear Admiral John E. Gingrich,** who resigned on May 3, 1949.

Jack W. Garrison, physicist at Armour Research Foundation of Illinois Institute of Technology, will head the newly organized nucleonics section in the Foundation's physics department.

Norris E. Bradbury, who has been Director of the Los Alamos Scientific Laboratory since 1945, has been appointed Professor of Physics at the University of California in Berkeley.

G. B. Arfken and **L. C. Biedenharn, Jr.,** have joined the theoretical physics staff at the Oak Ridge National Laboratories.

Marcus Oliphant is in Canberra to take up his new appointment as Director of the Research School of Physical Science of the Australian National University. Until the school is finally established, Professor Oliphant will be engaged in the construction and installation of the first of two cyclotrons planned for the school.

John R. Dunning, of Columbia University, and **Edward Mack, Jr.,** of Ohio State University, have been appointed to the Board of Directors of the Oak Ridge Institute of Nuclear Studies.

Donald W. Kerst, of the University of Illinois, has been awarded the John Price Wetherill Medal by The Franklin Institute for his "contribution to the theory of the betatron and the practical application of the theory."

Malvern J. Gross, formerly vice president in charge of engineering, General Electric X-Ray Corporation, has been appointed administrative assistant to Kenneth H. Kingdon, technical manager of the Knolls Atomic Power Laboratory, operated by General Electric in Schenectady, N. Y.

Sidney S. Minault has joined Tracerlab, Inc., as production manager.

D. M. Wroughton has been named manager of chemistry research at the Westinghouse Electric Corporation's Atomic Power Division. He succeeds **E. Bruce Ashcraft** who has been appointed advisory engineer in the chemical department of the Westinghouse Research Laboratories.

William A. Wildhack will head the Office of Basic Instrumentation, just established at the National Bureau of Standards. He was formerly chief of the Bureau's Missile Instrumentation Section.

IN BRIEF

➢A large plant has been established near the uranium mines at Buhovo, Bulgaria, for partial ore treatment. The "partial" treatment reduces ore volume to facilitate transport.

➢*Mexico's Institute for Scientific Investigation has signed a contract with the High Voltage Engineering Corporation, Cambridge, Mass., for the erection of nuclear research facilities.*

➢A U. S. Geological Survey investigation discloses the possibility that the Northern Peninsula of Michigan may contain deposits of radioactive materials. Airborne Geiger counters have located radioactive anomalies there.

➢*Improved methods of separating and preserving blood components will be sought in a program administered by the American National Red Cross under AEC contract. The ultimate purpose is to provide stockpiles of blood constituents for use in the event of atomic disaster.*

➢The board of directors for the American Society for Testing Materials has authorized the formation of a new technical committee on Sampling and Analysis of Atmospheric Pollution.

➢*Expansion of AEC's raw materials facilities will include construction of a new uranium ore purchasing station and processing plant for the autunite-torbenite deposits in Marysville, Utah.*

➢The danger of contaminating public water supplies by atomic waste material was discussed in a seminar at the University of California on Sept. 7–8.

➢*The Alco Products division of the American Locomotive Co. recently announced that it has begun work on a $3,000,000 order for nickel-plated pipe for the Atomic Energy Commission.*

MEETINGS

Institute of Radio Engineers and American Institute of Electrical Engineers Conference on Electronics Instrumentation in Nucleonics and Medicine—Park-Sheraton Hotel, New York, N. Y., Oct. 23–25

American Institute of Electrical Engineers, Fall General Meeting—Skirvin Hotel, Oklahoma City, Okla., Oct. 23–27

American Society for Metals, 32nd National Metals Congress and Exhibition—Chicago, Ill., Oct. 23–27

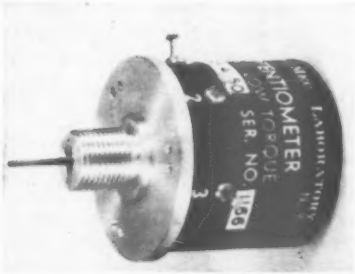
American Standards Association, 32d Annual Meeting—Waldorf-Astoria Hotel, New York, N. Y., Nov. 27–29

American Society of Mechanical Engineers, Annual Meeting—Hotel Statler, New York, N. Y., Nov. 27–Dec. 1

American Institute of Chemical Engineers, Annual Meeting—Columbus, Ohio, Dec. 3–6

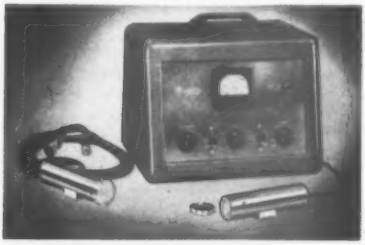
Radiological Society of North America, Inc., 36th Annual Meeting—Chicago, Ill., Dec. 10–15

PRODUCTS and MATERIALS



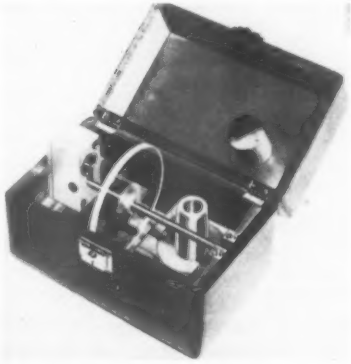
LOW-TORQUE POTENTIOMETER

Electro-Mec Laboratory, 225 Broadway, New York, N. Y. Requiring a shaft torque as low as 0.003 inch-ounces, this new potentiometer is available in models capable of carrying currents as high as 0.1 amp. Resistance values between 50 and 200,000 ohms can be provided. Accuracies as high as 0.05% are claimed. The instruments are supplied with ball, sapphire, or bronze bearings. Frictionless shaft seals protect the instrument against dust; insulation is treated with an antifungicide.



LABORATORY MONITOR

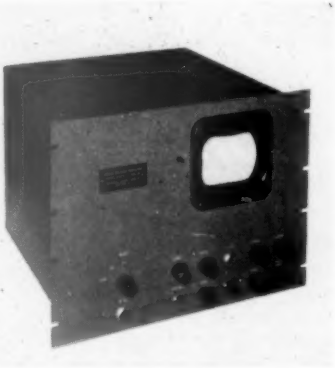
Nuclear Instrument & Chemical Corp., 229 W. Erie St., Chicago 10, Ill. The model 1615 monitor for alpha, beta, and gamma detection indicates activity in five ranges, from 500 to 50,000 cpm, on a count-rate meter. A switch on the instrument panel enables the operator to choose time constants which will give standard deviations of 3% or 20% on any range. The volume of the built-in speaker for aural monitoring is adjustable, as is the output of the high-voltage supply. Provision is made for attaching a chart-type recorder. A permanent magnet is incorporated in probes supplied with the monitor; this allows mounting of the probe on any ferromagnetic material.



Sr-90 MEDICAL APPLICATOR

Tracerlab, Inc., 130 High St., Boston 10, Mass. Utilizing a source of about 25 mc of Sr^{90} mounted at the end of a 6¾-in. shaft, the model RA-1 medical applicator is designed for use in the treatment of certain surface conditions. The source has an active diameter of 7.8 mm and an outside diameter of 12.7 mm. Protected by a double hermatic seal, the activity is covered with 2 mils of stainless steel and 10 mils

of aluminum, a total covering of about 100 mg/cm². It is claimed that essentially only beta radiation is given off by the source. A circular Plexiglas shield is mounted on the shaft for the protection of the operator.

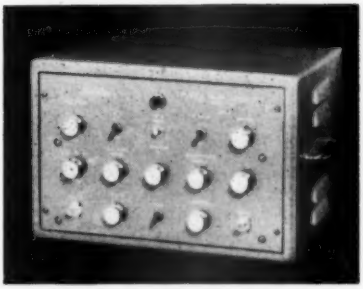


BRIDGE NULL INDICATOR

Industrial Control Co., 1452 Undercliff Ave., New York 52, N. Y. The model 203-A null indicator converts d-c bridge unbalance into an alternating voltage which is displayed on a built-in 5-in. cathode-ray tube. A 50-mv d-c signal is said to produce a 4-in. deflection on the screen. Gain is variable in decimal steps. Input resistance is greater than 2 megohms. Internal noise is claimed to be less than 5 μv and the instrument is said to be free of drift. The instrument operates on 110 volts a-c.

C-14 LABELED COMPOUNDS

Isotopes Division, U. S. AEC, Oak Ridge, Tenn. Several C^{14}-labeled compounds have recently become available for distribution through Oak Ridge National Laboratory. These compounds are as follows: uniformly labeled glucose and sucrose; 1,2-benzanthracene-5,6-C_1^{14}; chrysene-5,6-C_1^{14}; paraformaldehyde; and phenanthrene-9-C^{14}.



DOUBLE PULSE GENERATOR

Berkeley Scientific Co., 6th and Nevins Sts., Richmond, Calif. The model 902 double pulse generator produces two pulses individually controllable in width, amplitude, and time relation to each other. Pulse amplitude is adjustable from 0 to 50 and 0 to −200 volts without cross effect. Pulse duration is adjustable from 0.15 to 1.5 μsec. Spacing between the two pulses, with a rise time of 0.05 μsec and decay time of 0.10 μsec, is continuously variable in two ranges, −0.5 to 3 μsec. Repetition rate can be externally triggered; it is adjustable in three ranges: 1 to 10, 10 to 100, and 100 to 1,000 cycles. Output impedance is 400 ohms; maximum output voltage is −200 volts. Over-all calibration accuracy is ±5%.

REFRACTORY MATERIAL

Babcock & Wilcox Co., 85 Liberty St., New York 6, N. Y. Produced from a base of mullite, Allmul refractory material is available as a grain and a ramming mix as well as in firebrick. Its melting point is 3,335° F, and cold crushing strength is 1.7 tons/in². High resistance to thermal shock or spalling is claimed.

LEAD GLASS FABRIC

Bar-Ray Products, Inc., 209-25th St., Brooklyn, N. Y. Spun lead glass woven into a fabric is now available in 36-in. wide rolls. Gowns and aprons made

from the material are also available. It is claimed that the fabric provides ample protection of the body from X-radiation, and that it is highly resistant to beta radiation.

DRY WASTE CONTAINER

S. Blickman, Inc., 536 Gregory Ave., Weehawken, N. J. Designed for safe temporary storage of dry radioactive waste, this all stainless steel container is made up of an inner pail, outer pail, and cover assembly. A segment of the cover rotates when the foot lever is depressed; the segment closes the opening automatically when the lever is released. It is claimed that movement of the segment does not cause air disturbance which might blow waste out of the can. Pails have rounded corners and are free of crevices. The device holds 7½ gal of waste.

LABELED REFERENCE SHEETS

Isotopes Division, U. S. AEC, Oak Ridge, Tenn. Sheets of polystyrene labeled with C^{14} that can be used to prepare reference sources of weak beta radiation for instrument calibration are now available. Thickness of the sheets is about 0.010 to 0.012 in., somewhat greater than infinite thickness. Square sheets are available in two sizes: 1¼ × 1¼ in., weighing about 275 mg and containing 0.6 μc C^{14}; and 5 × 5 in., weighing about 4.4 gm and containing 9.6 μc C^{14}. Two sheets with the same dimensions will not necessarily have the same activity, nor will two sides of the same sheet necessarily give the same counting rate. With a 25-mm diameter source and an average G-M assembly, the activity of the material used is about 3,600 cpm. A single smaller sheet can be obtained without specific authorization by the AEC.

COUNT INDICATOR

Technitrol Engineering Co., 3211 Woodland Ave., Philadelphia 4, Pa. The model 137 radiation count indicator provides two channels for count-rate measurement and recording. Power supplies are provided for both scintillation and Geiger-tube probes. Total count is indicated on a chart recorder. With a 3-μsec resolution time, the instrument provides count rates of 100, 1,000, 10,000 and 50,000 cpm full scale. The standard counting time is 1 min; a built-in reset timer can be used to obtain variable counting times.

LITERATURE AVAILABLE

Direct Writing Recorders. Catalog sheets describe galvanometers, instrument amplifiers, and chart drives used with heat-sensitive paper. *Sanborn Co., 39 Osborn St., Cambridge 39, Mass.*

Van de Graaff. Bulletin A-S describes application of 2-Mev accelerator to research problems. *High Voltage Engineering Corp., 7 University Rd., Cambridge 38, Mass.*

pH Electrodes. Catalog EN-S5 details laboratory and industrial electrodes and assemblies for *p*H measurement. *Leeds & Northrup Co., 4907 Stenton Ave., Philadelphia 44, Pa.*